Floating Treatment Wetlands for Synthetic Greywater Remediation

By

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Abbreviation

Ag	Mercury
Al	Aluminium
As	Arsenic
AMD	Acid Minewater Drainage
ASCE	American Society of Civil Engineers
Avg.	Average
AWWA	American Water Works Association
B	Boron
BOD_5	Five–day Biochemical Oxygen Demand
BS	British Standard
BW	Black Water
C	Carbon
Ca	Calcium
Cd	Cadmium
CH_4	Methane gas
Cl	Chlorine
Co	Cobalt
CO_2	Carbon Dioxide
COD	Chemical Oxygen Demand
Cr	Chromium
Cu	Copper
CWs	Constructed Wetlands
DIC	Dissolved Inorganic Carbon
DO	Dissolved Oxygen
DOC	Dissolved Organic Carbon
DON	Dissolved Organic Nitrogen
DW	Distilled Water
Eh	Redox Potential
EPA	Environmental Protection Agency
EPE	Environmental Protection Agency
FAO	Food and Agricultural Organisation
Fe	Iron
FSF	Free Surface Flow
FTWs	Floating Treatment Wetlands

FWSCWs	Free Water Surface Constructed Wetlands
H	Hydrogen
HC	High Concentration Pollutants Synthetic Greywater
HCl	Hydrochloride Acid
HF	Horizontal–Flow
HRT	Hydraulic Residence Time
IBM–SPSS	International Business Machine Statistical Package for Social Sciences
ICP–OES	Inductively Coupled Plasma–Optical Emission Spectrometry
ISO	International Standard for Standardization
IWA	International Water Association
GW	Greywater
K	Potassium
LC	Low Concentration Pollutants Synthetic Greywater
Max	Maximum
Mg	Magnesium
Min	Minimum
Mn	Manganese
Mo	Molybdenum
MWTPs	Mine Water Treatment Plants
N	Nitrogen
n	Number of tested samples
na	Not applicable
Na	Sodium
NaOH	Sodium Hydroxide
NH_4–N	Ammonia–Nitrogen
Ni	Nickel
NOAA	National Oceanic and Atmospheric Administration
NO_2–N	Nitrite–Nitrogen
NO_3–N	Nitrate–Nitrogen
NTU	Nephelometric Turbidity Unit
O	Oxygen
OP	Organic Phosphorus
P	Phosphorus
Pb	Lead
Pe	Person equivalent
pH	Hydrogen ion
PIC	Particulate Inorganic Carbon

POC	Particulate Organic Carbon
PON	Particulate Organic Nitrogen
PO_4–P	Ortho–Phosphate–Phosphorus
PPCPs	Pharmaceutical and Personal Care Products
RZM	Root Zone Method
S	Sulphur
SC	South Carolina
SD	Standard Deviation
Se	Selenium
SF	Surface Flow
Si	Silicon
SGW	Synthetic Greywater
SS	Suspended Solids
SSF	Sub–Surface Flow
TBD	Turbidity
TDS	Total Dissolved Solids
TKN	Total Kjeldhal Nitrogen
TN	Total Nitrogen
TP	Total Phosphorus
TPFO	Toxicological Profile for Fuel Oils
TSS	Total Suspended Solids
USDA	United States Department of Agriculture
UK	United Kingdom
UN	United Nations
USA	United States of America
USEPA	United States Environmental Protection Agency
UV	Ultraviolet
VF	Vertical Flow
VFS	Vertical Flow Systems
VOCs	Volatile Organic Compounds (Carbon)
VPH	Volatile Petroleum Hydrocarbon
VSF	Vertical Sub–surface Flow
WW	Raw Wastewater
XOCs	Xenobiotic Organic Compounds
Zn	Zinc

A B S T R A C T

Aggravation of water scarcity, eutrophication and pollution, has led to treatment and/or recycling of the major fraction of domestic wastewater, which is grey wastewater, for non–potable options. The major aim of this study was to evaluate the performance of FTWs for remediation of various pollutants from synthetic greywater. In the present study, a novel synthetic greywater composition has been proposed with respect to the composition of organics, nutrients, heavy metals and other trace elements. As an imitation of bio–physiochemical treatment technology, mesocosm–scale retention ponds were artificially retrofitted with floating reedbed *Phragmites australis* to introduce, what is known as, floating treatment wetlands (FTWs) for improving the removal efficiency of soluble contaminations from synthetic greywater (SGW) in such systems. As a corroboration to the phytoremediation, pelletised ochre (acid mine wastewater sludge) was added into those FTWs for its capacity to adsorb phosphorus and other trace elements. Mainly, four operational variables were designated for the experimental design set–ups of the FTWs, namely: pollutant strength (high– (HC) and low– (LC) concentrations), contact time (2– and 7–day), presence of *P. australis* and presence of cement–ochre pellets. The findings revealed that dissolved (DO), and biochemical and chemical oxygen demands (BOD_5 and COD) decreased significantly ($p < 0.05$) in all FTWs. The concentrations of NO_3–N were significantly ($p < 0.05$) higher, and of PO_4–P were significantly ($p < 0.05$) lower than those concentrations of inflow, however, there were no significant ($p > 0.05$) changes in NH_4–N concentration in FTWs using a combination of ochre pellets with floating *P. australis*. Presence of ochre pellets in combination with *P. australis* significantly ($p < 0.05$) decreased Al, B, Cd, Cr, Cu, Mg, Ni and Zn, while increasing Al, Ca, Fe and K contents in the outflow. Accumulation of all considered elements was significantly ($p < 0.05$) higher in *P. australis* of all treatment systems (T) than in control systems (C), except for sodium (Na) contents which had significantly ($p < 0.05$) lower accumulations. The statistical analysis showed that accumulated concentrations of all considered elements in ochre pellets after the end of the treatment experiment were significantly ($p < 0.05$) higher, except for Ca concentrations which were significantly ($p < 0.05$) lower, than those in ochre pellets before commencing the experiment.

Chapter One

Introduction

1.1. Background

Water crisis is considered a crucial problem internationally due to its relationship with health, economy of communities and the environment. In the last century, the demand for freshwater increased by double that of the population growth rate (UN–Water, 2006).

For the world's population, agriculture is the main source of food followed by biodiversity sources. The total irrigated agriculture required about 70% of available water sources. This percentage is expected to increase by around 14% by 2030 (FAO, 2003). Concurrently with population rising, freshwater quality tends to suffer degradation due to people and their dietary needs, fuel production, aged infrastructure, pollution, and unsustainable disposal practices of agricultural and industrial wastes (FAO, 2011). Coupled with climate change, such as insufficient rainfall and high temperature, it is expected that the global water shortage will increase by about 20%, and one–third of the world could be at the limit of sufficient water by 2025. Furthermore, the expectation could be worse in some countries which will be living under absolute water stress (Pereira et al., 2009).

In fact, industrial development and energy generation are also contributing to increase the pressure on natural water sources associated with the production of large volumes of wastewater (USEPA, 2012). On the other hand, the balance between freshwater withdrawal and wastewater disposal, from various resources, has emerged as a critical emphasis for sustainability of water sources (USEPA, 2004). This issue has driven conservation of water sources in two directions. Firstly, by considering alternative

resources of water, such as deeper exploitation of groundwater, and seawater desalination, as well as constructed reservoirs and dams to maintain rain water. But the expected costs of those strategies are greater than those of conventional water sources and they may have negative impacts on the environment (Friedler, 2004).

Secondly, a suggestion has been made to reuse or recycle wastewater through the promotion of sustainable management techniques where all types of wastewater must be treated to a specified standard before being discharged into water bodies, such as oceans and rivers (USEPA, 2012; El–Kady and Suloma, 2013).

1.2. Justification and Motivation

From environmental and economic aspects, both industrial and developing communities are interested in exploring alternative unconventional water resources by diversion of wastewater for non–drinking uses to mitigate water scarcity, surface water pollution, and treatment cost (USEPA, 2012; Kanarek et al., 1993). However, there are a number of complexities associated with wastewater reuse, regarding the requirements, treatment approaches, regulations and limitations to minimise the community's health and environmental risks (Dalahmeh and Baresel, 2014). In contrast, short–term and high–cost strategies to meet water demands (e.g. exploration of deep groundwater and seawater desalination) are likely to have detrimental effects on the environment, such as ground subsidence, salinity intrusion, and ecosystem deterioration (Friedler, 2004; D. Zhang et al., 2014). Although it is expensive to supply water at sufficient quality for human consumption, drinking water continues to be used for non–potable applications (Eriksson, et al., 2002).

The concept of treating and reusing urban wastewater is not new. Wastewater is the most reliable supply for reuse for all possibilities, since it is the only resource that

increases along with population. It is common practise to recycle wastewater for agricultural irrigation, in particular for arid and semi–arid regions (Jefferson et al., 1999). Irrigating residential properties, golf courses, public parks, crops, flowers and forests are among the available options. It can also be used for toilet and urinal flushing in residential and commercial buildings (Friedler and Galil, 2003). Entertainment landscaping can also be served by recycling water in water features such as fountains, pools and waterfalls. Industrial sectors reuse water for applications including air–conditioning, cooling, general wash down and street cleaning. Fire protection is another potential use option, as is the use of reclaimed water in construction activities such as making concrete (Eriksson et al., 2002). The wastewater reuse options mentioned so far are often fairly large–scale operations undertaken by local authorities (Emmerson, 1998).

Domestic wastewater generally consists partially of blackwater, containing faecal discharge of toilets, with the majority bring greywater which includes washing wastewater from showers, dishwashers, basins, and laundry (Christova–Boal et al., 1996; Jeppesen, 1996). Due to the fact that greywater constitutes 50–80% of the total household wastewater (Eriksson et al., 2002) and to the low levels of contaminating pathogens and nitrogen, reuse and recycling of greywater is receiving more and more attention (Al–Jayyousi, 2003). It is now widely accepted that greywater recycling is feasible and can contribute to sustainable water management (Domènecha and Saurí, 2010). However, greywater–only schemes are currently the poor relations of water recycling activities on the global stage (Surendran, Wheatley, 1998; Pidou et al., 2007).

Recycling wastewater should fulfil the criteria of hygiene, safety, aesthetics, environmental tolerance, and technical and economic feasibility (Nolde, 1999; Anda, et al., 2002), otherwise nutrients and other pollutants will threaten the surface water and groundwater (Shin et al., 1998). Nutrients such as nitrogen and phosphorus are mainly

responsible for water eutrophication from diffuse or non–point sources associated with surface runoff and from point sources typically associated with concentrated farming activities such as the production of livestock, and industrial, and agricultural runoff (Scholz, 2003; Garland et al., 2004; Dunne et al., 2012). Environment and human health problems might be associated with eutrophication such as excessive growth of phytoplankton, including algae, resulting in toxin excretion, oxygen depletion, stench, and aquatic animals' mortality (Scholz, 2006; Wei et al., 2008).

To control eutrophication, all liquid wastes must be treated to a specified standard before being discharged into a water body (SEPA, 2002; Dzakpasu et al, 2012). Treatment of generated wastewater is a major dilemma for governments regarding the environment. The traditional strategy for sewage management has been devised according to minimum treatment and discharge into surface water bodies.

The generated volume of waste requiring treatment is a major problem for water authorities and the environment. The traditional response to sewage management has been a system based on minimal treatment and discharge into receiving waters, such as oceans and rivers. This has further given the impetus to the development of alternative wastewater treatment techniques that meet regulatory criteria, whilst also providing a more environmentally sustainable option.

Among the different treatment systems now available worldwide, the use of ecological technologies such as constructed treatment wetlands has attracted more attention, leading to the evolution of constructed treatment wetlands as a viable alternative to traditional wastewater treatment systems (Jefferson et al., 1999). Furthermore, they represent an innovative and emerging solution for environmental protection and restoration, largely because of their moderate capital costs, very low

energy consumption and low maintenance requirements, and benefits of increased wildlife habitat (Scholz and Lee, 2005; Vymazal, 2007; Teng et al., 2012). Constructed wetlands are engineered wastewater treatment systems that encompass a plurality of treatment modules including biological, chemical, and physical processes akin to processes occurring in natural treatment wetlands (Brix, 1994; Vymazal, 2005; Scholz, 2010). Indeed, constructed wetlands have proved to be efficient in removing the main chemicals (organic substances, heavy metals, etc.) and biological organisms (bacteria, viruses, parasites, etc.) from municipal and domestic wastewaters (Gross et al., 2007a). One of the main biological components of wetlands are macrophytes, which not only assimilate pollutants directly into their tissues, but also act as catalysts for purification reactions, increasing environmental diversity in the rhizosphere and promoting a variety of chemical and biological reactions (Tchobanoglous, 1993).

On the other hand, wetland vegetation provides removal mechanisms for soluble pollutants or finer particles but usually requires larger areas and shows limited tolerance for extended periods of high water levels. With existing wetland technology, it is possible to treat the water in sewage to levels required for any of the current applications for water.

1.3. Floating Treatment Wetlands (FTWs)

In general, the various types of constructed wetlands require relatively simple engineering efforts with additional lands resulting in extra costs for the treatment budget. Therefore, floating treatment wetlands are considered as an innovative technology for water purification which could be more economical than conventional constructed wetlands. They might be classified as either natural or artificial, both being a combination of pond and floating vegetation in a hydroponic manner (Billore, 2007; Billore et al., 2008; Tanner and Headley, 2008; Tanner and Headley, 2011). The colonised roots of

macrophytes immerse into the water column filtering suspended contamination and nutrients from the water (Brix, 1997; Stewart et al., 2008; Wen and Recknagel, 2002). Furthermore, these roots provide oxygen into the rhizosphere for the substrate, for growth of a biofilm of microorganisms which breakdown the organic substances as well (Brix, 1987; Nakamura and Mueller, 2008; X. Li et al., 2009; Headley and Tanner, 2012).

The aerial parts of the vegetation could also serve as a habitat and food for biodiversity, and for aesthetic aspects (Knight, 1997; McNally and Wilkinson, 2011). In addition, macrophyte biomass such as *P. australis* in wetlands enhances filtration and sedimentation, mitigating the possibility of resuspension of settled solids by slowing water flow velocity down and increasing the contact time between wastewater and substrate (Vymazal, 2011; Leto et al., 2013). Furthermore, macrophytes change the physicochemical conditions in the water column by releasing bioactive compounds from the plant roots, which might enhance sorption and/or sedimentation processes in the vegetated FTW (Tanner and Headley, 2011).

The design of floating treatment wetlands is a simulation of those which are natural, but adding an artificial buoyant mat enables wide species of macrophytes to grow (Mallison et al., 2001; Faulwetter et al., 2011). Floating treatment wetlands have been widely used for pollutant removal, such as organics, nutrients, and metals from surface water, storm runoff, agriculture runoff, mine drainage, and industrial, and municipal wastewater (Hammer, 1992; Naveh, 1998; Akcil and Koldas, 2006; DeLorenzo and Fulton, 2009; John et al., 2009; Hubbard, 2010; Gallagher et al., 2011).

Among the various removal mechanisms in FTWs, sedimentation seems to be the principle process of pollutant removal compared to other processes such as macrophyte uptake (Gill et al., 2014; Borne et al., 2015). Oxygen demand removal may be attributed

to the settling of suspended solids and trapping of them into the plant rhizomes and roots of constructed wetlands, as well as biological degradation (Zhang et al., 2015).

1.4. Acid Minewater Drainage (AMD)

There has been a wide interest in recycling and reuse of industrial waste sludge, such as ochre, for remediation of various types of wastewater, particularly to adsorb phosphorus (Heal et al., 2003). Ochre is a by–product of mineral sludge with a majority of hydrated ferric oxide ($Fe(OH)_3$) obtained from the drainage of disused coal mine works (Hancock, 2005). Ochre consists predominantly of the same hydroxides, such as aluminium (e.g., alum) or iron salts (e.g., ferric chloride), applied in traditional wastewater treatment processes for the removal of phosphorus (Sibrell et al., 2009). Furthermore, the cost of the chemical reagents used for the treatment of wastewater are relatively high. Both disposal and stockpiling of ochre sludge is costly. The annually generated ochre in the United Kingdom (UK) is around 32,000 tonnes. In comparison, the cumulative sludge in England is only about 19,000 tonnes/annum (Hancock, 2005; Dobbie et al., 2009). Thus, offsetting costs by exploring the feasibility of reusing ochre to improve wastewater quality is highly attractive.

Among various wastewater treatment technologies, constructed wetlands have been considered as a suitable best management practice for treating wastewater economically (low construction, operational and maintenance costs), energy–efficiently (natural physical, chemical and biological–based processes) and in an environmentally–friendly manner, compared to other more traditional civil and mechanical engineering approaches. However, the removal of phosphorus in constructed wetlands is not efficient, because of the deficiency in terms of adsorbing and retaining this element (Scholz and Lee, 2005). Wetlands are also often low in aluminium (Al) and iron (Fe). Although

phosphorus is vital for the growth of aquatic plants and microorganisms, too much leads to eutrophication of natural watercourses. Thus, the utilisation of ochre is highly recommended to increase the phosphorus removal in wetland systems as an alternative source of iron and aluminium oxides (Heal et al., 2003; Sibrell et al., 2009; Dobbie et al., 2009).

Because of its fine–grained texture and low hydraulic conductivity, ochre is unsuitable for use as a substrate for filter units or as wetland media. Difficulties, such as rapid clogging of treatment systems, when using raw ochre sludge have led to the creation of ochre in the form of granular pellets by mixing it with Portland cement (Heal et al., 2005; Dobbie et al., 2009; Sibrell et al., 2009; Carr 2012; Littler, 2012). Cement–ochre pellets are robust solids, easy–to–handle, linked to a high hydraulic conductivity and characterised by a high absorbent reactivity compared to raw ochre sludge. According to Littler et al. (2013), pelletised ochre was found to remove phosphorus in tests up to seven days. The pellets are characterised by high removal capacities and operational lifetime. In comparison, the unbound mine sludge seemed to reach its equilibrium within a phosphate solution after about 60 minutes of contact time (Littler et al., 2013). Pollutant removal from wastewater in the presence of cement–bound ochre may occur by physical precipitation under alkaline conditions, although this mechanism is considered less effective than removal by chemical sorption (Heal et al., 2005).

Phosphorus is present in domestic greywater, which is generated from household washing activities, at varying concentration levels due to the use of a wide range of detergents (Revitt et al., 2011). So, the use of ochre in wetlands for greywater treatment is an option (Gross et al., 2007b).

The research is significant, because it encourages chemical engineers to put a technique, which is novel in chemical process engineering, into practice. Furthermore, the research interprets rare detailed findings from a carefully executed experiment. Although ochre pellets within constructed wetlands are likely to improve the treatment efficiency of phosphorus, the fate of trace elements during the treatment process is uncertain. Therefore, this contribution focuses particularly on the fate of metals during remediation processes, which is novel in sustainable wastewater treatment.

1.5. Rationale, Aim and Objectives

There is a need to develop standard synthetic greywater recipes to allow for the easy comparison of similar experiments in the future. Original experiments and a detailed literature review have been performed to support the development of reasonably stable generic synthetic greywater recipes for both low and high pollutant concentrations. In this study, mesocosm–scale floating treatment wetlands (FTW) vegetated with *P. australis* were investigated to treat artificially prepared greywater. The first hypothesis of this study is that macrophytes are responsible for the removal of various pollutants. Sedimentation mechanisms may be ignored due to agitation of the outflow at the time of sampling. The second hypothesis was that *P. australis* is restricted in its natural activities when its rhizomes are subjected directly to contaminated greywater. However, this potential disadvantage had to be balanced with reduced land requirements and saving of aggregate resources.

The research also contributes to environmental chemistry and process engineering knowledge and understanding by promoting an overlooked opportunity for wastewater treatment and industrial waste recycling using environmental and chemical processes. The outlined methodology taken from environmental engineering applications controls

pollution and promotes resource recovery using clean processes that can be integrated into chemical engineering practice. Wastewater and waste are both minimised.

To date, floating treatment wetlands have not yet been used for treatment of either domestic or synthetic greywater. Therefore, the main aim of this research is the investigation of treatment of an artificially prepared greywater in simulated floating reedbed wetlands vegetated with *P. australis* (common reeds) in combination with cement–ochre pellets which examines their ability for removal of phosphate and other trace elements. So, the main objectives of this research are:

- Objective 1. To propose practical recipes to be used for the simulation of greywater, which can be used with confidence to assess different treatment technologies. This is achieved by the following steps:
    a) Review previous greywater recipes and corresponding components,
    b) Evaluate the quality of the new synthetic greywater and compare it with recipes found in the literature,
    c) Compare the quality of inflow synthetic greywater with outflow after 2– and 7–day contact time in treatment systems, and
    d) Prove that artificial greywaters act as real greywater under the effect of operational variables of the experimental set–up design.
- Objective 2. To assess the treatment performance of various experimental designs as a function of presence of macrophytes, increasing contact time of treatment by vegetated floating wetlands, and different concentrations of pollutant loads into the inflow. This is achieved by the following steps:
    a) Compare the outflow water quality with that of the inflow,
    b) Evaluate the effects of macrophytes on outflow water quality,
    c) Study the effect of contact time on treatment behaviour, and

d) Investigate the effect of inflow pollutant loads on the treatment system.

- Objective 3. To treat the artificial greywater with recycled waste by utilising ochre as pellets for the remediation of various pollutants including phosphorus and metals from contaminated greywater. This leads to the opportunity for resource recovery by extraction of concentrated elements from exhausted ochre pellets at the end of their lifetime in the future. This is achieved by the following steps:
    a) Identify the chemical composition of ochre and the possibility of trace element release from raw ochre and cement–ochre pellets into deionised water,
    b) Determine the pollutant removal (mainly phosphorus) from water in mesocosm–scale experiments under stagnant water conditions in stabilisation ponds, and
    c) Investigate the trace element accumulation into cement–ochre pellets after the end of the experiment.
- Objective 4. To evaluate the impact of *P. australis* in combination with cement–ochre pellets on removal of organic substances, nutrients, and metal pollutants from synthetic greywater as a plant–based remediation technology (phytoremediation). In other words, to evaluate the contribution of cement–ochre solids for water quality improvement with and without the presence of *P. australis*. This is achieved by the following steps:
    a) Evaluate the outflow water quality according to the operational parameters in experimental set–up design of wetlands such as treatment contact time, pollutant strength, presence of *P. australis*, and contribution of cement–ochre solids in treatment processes,

b) Evaluate and analyse cement–ochre pellets for accumulative heavy metals during the treatment period, and,

c) Evaluate and analyse biomass tissues of *P. australis* for accumulative heavy metals and their distribution in roots and rhizomes, stems, and leaves.

1.6. Research Procedures and Strategies

In the initial stage of this research, previous publications were reviewed to understand the circumstances related to treatment, recycling and/or reuse of domestic wastewater, greywater in particular. The focus was on treatment by constructed wetlands and floating treatment wetlands. In more specific detail, the role of macrophytes in wetlands is not yet clearly understood in treatment processes. However, some studies have claimed an efficient treatment of wastewater by wetlands with the presence of vegetation in removal of organic matter, nutrients, and metals. The characteristics of greywater have wide variation in both consistency and quality. So, synthetic greywater was suggested in this study to operate floating mats of *P. australis*. The experiment was operated on 1st September 2014 under real weather conditions and left for two months for biofilm development, until 1st November 2014 when cement–ochre pellets were used in the experiment to enhance the removal of phosphorus and heavy metals from the synthetic greywater. On 15th January 2015, the foliage parts of *P. australis* were harvested, dried, weighed, and stored for additional future tests. Furthermore, the same procedure was followed at the end of the experiment, November 2016. Through the treatment periods water quality was tested to assess treatment mechanisms and efficiency as a function of contact time, contamination loading rates, and existence of *P. australis*.

Artificial greywater, simulation of floating reedbed wetlands, cement–ochre pellets, set–up design of experiment, and different laboratory tests were prepared to

assess the role of floating mats of *P. australis* and ochre pellets in the treatment of synthetic greywater and its reuse for irrigation. Water quality sampling was carried out to monitor treatment performance variations. Ortho–phosphate–phosphorus and trace element removals were frequently low in the floating wetlands. Therefore, ochre and iron(III) oxide, have been used as cement–ochre pellets in the experiment.

Biomass tissues of *P. australis*, for two growth seasons, were analysed for evaluation of the accumulative heavy metals and their distribution into different locations, such as roots and rhizomes, stems, and leaves. Cement–ochre solids were also analysed for adsorption capacity of metal element concentrations before and after commencing treatment. All digested samples were prepared for analyses using Inductively Coupled Plasma–Optical Emission Spectrometry (ICP–OES) to indicate the accumulated trace elements.

1.7. Thesis Outline

This experimental work deals with treatment of the suggested synthetic greywater, using *P. australis* as a simulation of floating treatment wetlands. This research is described in eight chapters. Background on presented water shortage and the motivation of selected subjects and justifications for research are highlighted. Main aims and objectives to achieve those aims, materials and methods for carrying out the research are described in detail. Results and their discussion, and finally, the summarised conclusions and recommendations for future works are also included.

Chapter one is entitled “introduction” and contains seven sections. The background of the worldwide water scarcity and its effects on communities’ health and the nature are presented. The justification and motivation for investigating the potential of the suggested solutions are provided by the current and predicted future water crises.

An introduction is given into floating treatment wetlands (FTWs) and sludge (ochre) produced from acid minewater drainage (AMD). Then, rationale, aims and objectives are detailed. A brief review of the research strategies and thesis structure are presented at the end of this chapter.

Chapter two is designated for reviewing the published research linked to the thesis topic, starting with an overview and definition of greywater as a fraction of domestic wastewater followed by its volume, quality variation and published characteristics. The justifications for recycling and reuse of the treated greywater are illustrated regarding level of contaminates, such as organic matters, nutrient, metals and pathogens. Due to the variation in quality of real greywater worldwide, synthetic greywater was introduced in the operation of different treatment approaches, in previous literature. The recipes and corresponding greywater quality of produced greywater are also discussed. Chemical materials used either in household commercial products and/or for creating artificial wastewater are described and discussed. The reviewed publications also include information concerning the advantages of floating wetlands in treatment of different types of wastewater. Furthermore, using of industrial by–product wastes for aquatic treatment systems, such as minewater treatment sludge (ochre), are also reviewed.

Materials and methods are demonstrated in the third chapter. The experiment set–up design, chemical materials, recipes and preparation of synthetic greywater are illustrated in this chapter. Moreover, the water quality and the statistical data analysis tests, types and method, are also included. The protocol for creating cement–ochre pellets and the experimental scheme of their use within the floating treatment wetlands are also illustrated. The protocol for preparation of plant tissue samples for digestion to analyse for trace element concentrations is also described.

Results and their discussions were broken down into four chapters to distinguish the achievement of the proposed aims. So, chapter four shows the investigation of the chemical stability of simulated greywater as a function of time through observation of the water quality after 2– and 7–day retention times compared with synthetic greywater quality at the inflow stage, for both low and high pollutant concentrations of greywater.

In chapter five, mesocosm wetlands containing only floating *P. australis* are considered to study the effect of the presence of macrophytes on treatment performance compared with outflow quality of mesocosm wetlands containing only synthetic greywater. Furthermore, the interaction effects between the set–up design operational variable parameters on treatment efficiency are investigated as well.

Chapter six focuses on the role of cement–ochre pellets in removal performance of pollutants, mainly phosphorus and trace elements, from the synthetic greywater. The outflow greywater quality from mesocosms containing only ochre pellets are considered in comparison with effluents from systems containing only greywater. Moreover, the ochre pellets adsorption capacity and operational parameters effects are highlighted.

Chapter seven describes the performance of floating treatment wetland systems in combination with cement–ochre pellets in terms of outflow water quality, concentrations of trace element accumulated in the ochre pellets and in different parts of *P. australis* tissues, namely roots and rhizomes, stems and leaves.

According to the analysed results, chapter eight contains conclusions and suggested recommendations for future works. References are also included at the end of present research.

Chapter Two

Literature Review

2.1. Overview

In this chapter, grey wastewater is reviewed in terms of its definition, generated volume, composition and quality, as reported in previous studies. The physicochemical characteristics, trace element concentrations, presence of microorganisms, and expected effects of household chemicals are summarised. Due to the fact that greywater has a large variation in terms of quality, numerous publications have introduced different formulations for creating synthetic greywaters to mimic their associated substances, which are shown below. The common analytical grade chemicals used in simulation of artificial wastewater are listed and discussed. Those formulas have been utilised to operate a wide range of treatment approaches; one of which is constructed wetlands. Historical development of constructed wetlands (CWs) and their classification, according to their water flow path and level, have been reviewed. Furthermore, the removal mechanisms of different pollutants in wetlands have been highlighted. A review on floating treatment wetlands is also included to show the efficiency of floating macrophytes in treatment of various types of wastewater. The review includes the floating artificial mats which are cultivated with emergent, floating leaves, or free floating macrophytes within ponds. Additionally, the sustainable recycling strategy of industrial solid wastes such as ochre sludge, which presented on of acid minewater treatment sites, and its potential to remediate contaminated water are illustrated. Moreover, recycling of reclaimed wastewater for non–potable purposes, such as agricultural irrigation, is also mentioned in this chapter.

2.2. Greywater

According to the sources originated, household wastewater could include two types of discharge, namely Black and Grey wastewaters. Blackwater (BW) is defined as the derived effluents from toilet, bidets and urinals, while Greywater (GW) refers to untreated domestic wastewater produced from bathtubs, showers, hand basins, laundry machines, dishwashers and kitchen sinks, in households, office buildings, schools, etc. (Jeppesen, 1996; Jefferson et al., 1999; Eriksson et al., 2002). Occasionally, effluents generated from kitchen sinks, garbage disposal units or dishwashers are excluded from greywater (Christova–Boal et al., 1996; Emmerson, 1998; Al–Jayyousi, 2003; WHO, 2006b), as shown in Figure 2.1.

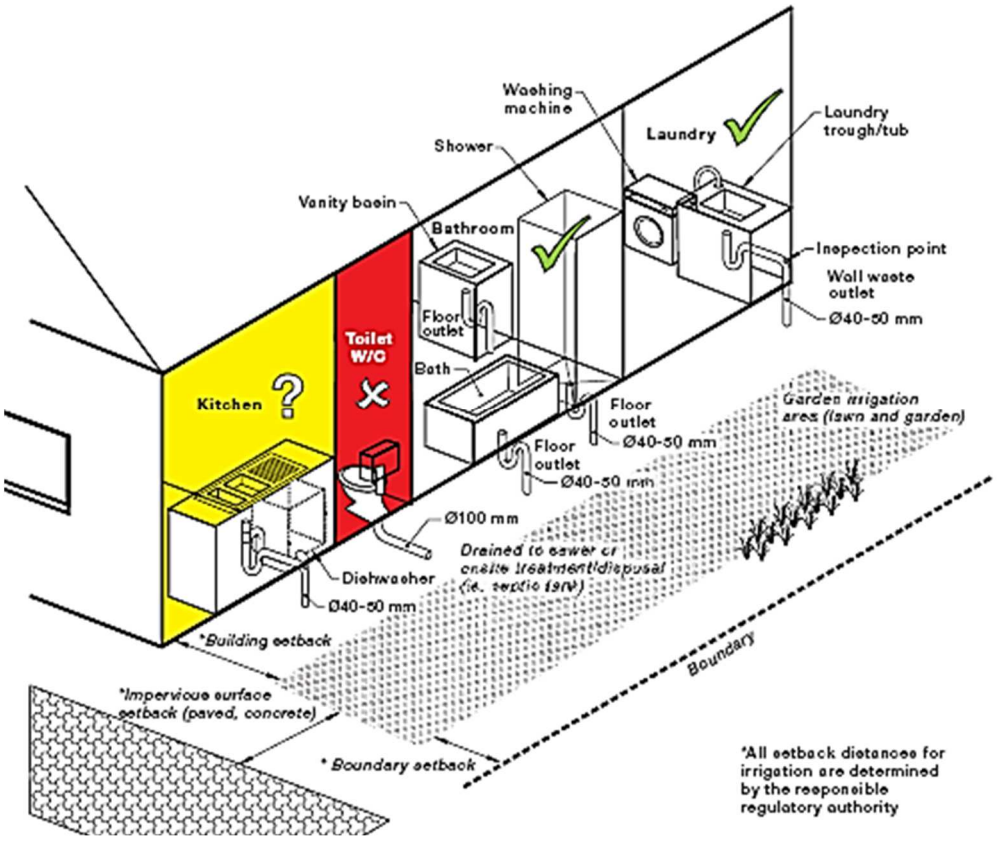


Figure 2.1. Sources of domestic greywater (ANWC, 2008).

Volume and quality of greywater vary from source to source and in terms of both place and time due to the variations in water consumption in relation to the discharged amounts of substances. This variation mainly depends on lifestyles, living requirements, demography (age, gender), customs and habits, water installations and the degree of water abundance (Eriksson et al., 2002; Friedler et al., 2005). Furthermore, the quality of water supply could also essentially affect greywater properties. Leaching from piping and biological degradation processes in the biofilm attached to network piping walls for both distribution of drinking water and collection of greywater, through transporting or storing, might also have an effect on greywater properties.

Furthermore, greywater is likely to be the major contributor to the total wastewater quantity generated from an average household (Figure 2.2). It represents the largest potential source of water savings in domestic residences, accounting for as much as 50–80% of the total water used (Al–Jayyousi, 2003; Schäfer et al., 2006; F. Li et al., 2009). In general, it consists of a small fraction (no more than 0.5%) of solid wastes, while the major portion is used water (Emmerson, 1998). Due to its large amount, low organic content (about 30%), and small fraction of nutrients (9–20%) (Pidou et al., 2007), greywater could be recycled and added to the water resources as a significant contribution to water savings for domestic residences (Al–Jayyousi, 2003; Friedler et al., 2005).

Hygiene, safety, aesthetics, environmental tolerance and economic feasibility are the most important standard criteria that the recycled greywater needs to fulfil for reuse (Nolde, 1999). Inadequately treated or disinfected greywater presents a risk of infection to end users from pathogens in the reused greywater (Jeppesen, 1996). Historically, the greywater constituents are similar to those of domestic wastewater (Christova–Boal et al., 1996; Eriksson et al., 2002; F. Li et al., 2009). However, greywater usually contains far fewer pathogens and, hence, is much more suitable (and socially more acceptable) for

recycling options in a single household context (Al–Jayyousi, 2003; Pidou et al., 2007; Domènech and Saurí, 2010).

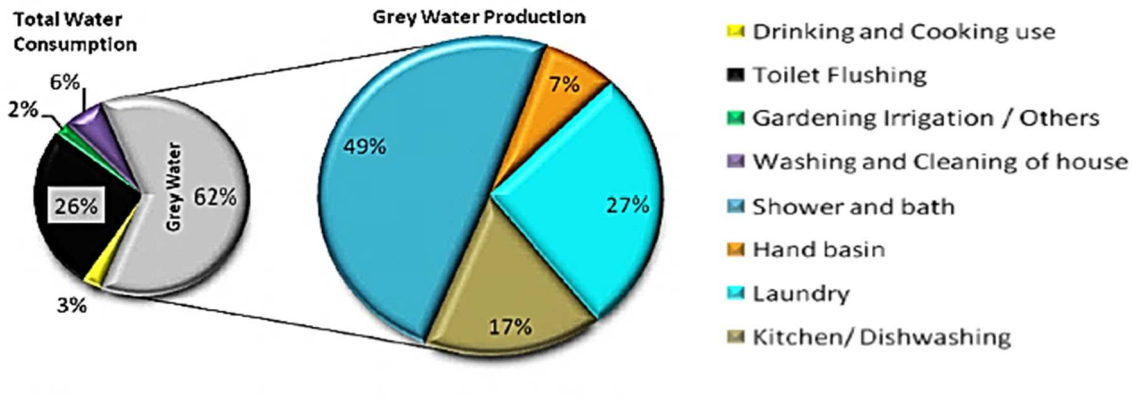


Figure 2.2. The relative distribution of total water consumption and greywater production (Edwin et al., 2014).

The most common non–potable applications of wastewater reuse, particularly greywater, are in agricultural (Gross et al., 2014), industrial, urban and environmental sectors (Surendran and Wheatley, 1998). The water that is used for toilet flushing in many countries today is of drinking water quality (Nolde, 1999; Eriksson et al., 2002; Mourad et al, 2011).

Due to the low levels of contaminating pathogens and nitrogen content (Benami et al., 2015), reuse and recycling of greywater have gained special interest (F. Li et al., 2009). Thus, on–site irrigation is another application for grey wastewater reuse, and is becoming common practice worldwide (Santos et al., 2014). It is used for irrigation of lawns, athletic fields, cemeteries, parks and golf courses, as well as domestic gardens. Washing of vehicles and windows, fire protection, boiler feed water and concrete productions are examples of other suggested on–site usages (Pidou et al., 2008). In addition, grey wastewater could be used to develop and preserve wetlands. An alternative way of handling grey wastewater is to infiltrate it into the ground and thereby create a shortcut in the urban hydrological cycle.

Investigation into the impact of grey wastewater storage found that storage for 24 hours improved the quality of the water but storage for more than 48 hours could be a serious problem as the dissolved oxygen might be depleted (Eriksson et al., 2002). The 5–day biochemical oxygen demand (BOD_5) and dissolved oxygen (DO) concentrations decrease during the sedimentation period when greywater is stored. Evidence has shown that 50% removal of BOD_5 could be achieved when greywater is stored over a four–hour period (Jefferson et al., 1999). However, extended storage may lead to the risk of odour increases and possibly health issues, due to enhanced microorganism growth (Jefferson et al., 2001). Furthermore, the BOD_5 concentration, for example, in greywater originating from hand–washing basins has been reported as being slightly lower and more variable than the one generated from mixed resources, in addition it varies with different discharge patterns (Al–Jayyousi, 2003). There has been considerable research into the quality processes of raw greywater occurring during the storage stage (Liu et al., 2010). For example, Dixon et al. (1999) indicated improvements in greywater quality during complex storage processes. Focusing on literature, a variation in data has been noticed not only between sources but also between locations for the same source. It has been highly recommended to assess and evaluate the quality of greywater before reuse and treat it if needed. Published characteristics of different types of greywater according to source of generation are summarised in Tables 2.1–2.4.

Table 2.1. Characteristics of real greywater.

Reference	Greywater source	Temp. (°C)	pH (–)	Turbidity (NTU)	TSS (mg/l)	EC (µS/cm)	DO (mg/l)	BOD_5 (mg/l)	COD (mg/l)	NH_4–N (mg/l)	NO_3–N (mg/l)	PO_4–P (mg/l)
Eriksson et al. (2002)	Bathroom GW	29	6.4–8.1	60–240	54–200	82–250		76–200	100–633	≤0.1 to 15.0	0.28–6.30	0.94–48.80
	Laundry GW	28–32	8.1–10.0	14–296	120–280	190–1400		48–380	12.8–725.0	0.04–11.30	0.4–2.0	4–171
	Kitchen GW	27–38	6.3–7.4		235–720		2.2–5.8	1040–1460	3.8–1380	0.002–23.0	0.3–5.8	12.7–32.0
	Mixed GW	18–38	5.0–8.7	15.3 to ≥200.0		320–20000		90–360	13–549	0.03–25.40	0.0–4.9	4–68
Al–Jayyousi (2003)	Hand basin							109	263	9.6		
	Combined			69				121	371	1		
	Single person			14				110	256			
	Single family			76.5						0.74		
	Block of flats			20				33	40	10		
	College			59				80	146	10		
	Large college			57				96	168	0.8		
Christova–Boal et al. (1996)	Bathroom GW	25	6.4–8.1	60–240	48–120	82–250		76–200		≤0.1 to 15.0		
	Laundry GW	25	9.3–10.0	50–210	88–250	190–1400		48–290		≤0.1 to 1.9		
F. Li et al. (2009)	Bathroom GW		6.4–8.1	44–375	7–505			50–300	100–633			
	Laundry GW		7.1–10.0	50–444	68–465			48–472	231–2950			
	Kitchen GW		5.9–7.4	298	134–1300			536–1460	26–2050			
	Mixed GW		6.3–8.1	29–375	25–183			47–466	100–700			
Al–Hamaiedeh and Bino (2010)	Real GW range		6.9–7.8		23–358	157–200		110–1240	92–2263		0.44–0.93	
	Real GW average		7.2		275	183		942	1712		0.68	
Pidou et al. (2008)	Mixed GW LC		6.6–7.6	35				39	144	0.7	3.9	0.5
	Shower GW HC		7.3–7.8	42				166	575	1	7.5	1.3
	Real Raw GW			46.6				205	791	1.2	6.7	1.66
Ramona et al. (2004)	Shower GW1		7.5	23	29.8	1317		78	170	1.5–3.0	0.05–1.70	0.02–0.19
March et al. (2004)	Raw GW		7.3–8.0	5–62					39–441			
Eriksson et al. (2010)	Raw GW1		7.7–8.1		51–135		2.5–4.5	18–68		0.36–4.40		0.02–2.20
	Raw GW2		8.2–8.3		67–390		9.3–9.5	≤3		0.07–0.13		0.25–0.28
Nghiem et al. (2006)	Real GW		5.0–10.9					33–1460	3.8–1380.0			
Houshia et al. (2012)	Raw GW		6.1			1500		126.6			38	
Leal et al. (2012)	Raw GW		7.24			74.4			1476		≤0.10	2.97

Note: pH, hydrogen ion; TSS, total suspended solids; EC, electrical conductivity; DO, dissolved oxygen; BOD_5, 5–day biochemical oxygen demand; COD, chemical oxygen demand, NH_4–N, ammonia–nitrogen, NO_3–N, nitrate–nitrogen; PO_4–P, ortho–phosphate–phosphorus and NTU: Nephelometric Turbidity Unit.

2.2.1. Physical Properties

The relevant physical properties of greywater are colour, temperature, turbidity, and total suspended solids. In general, colour of greywater varies from source to source depending on the household habits and uses, however, the values seem to be absent or neglected in most publications, (Table 2.1). Temperature also varies and is related to geographic location and purpose of water uses, for example, general hygiene needs warm water. Moreover, the relative high temperature may stimulate microorganism growth and breeding (Rose et al., 1991; Winward et al., 2008). Elevated warming of supersaturated greywater can also contribute to the presence of calcium carbonate and some inorganic salts, since their solubility in water decreases with elevated temperatures (Shin et al., 1998; Nghiem et al., 2006). The reported temperature values were in the range 18–38 °C (Santala et al., 1998; Erikson et al., 2002).

Colloids and particles can be indicated by turbidity and total suspended solids measurements. Colloids are non–crystalline, homogeneous substances of large molecules or ultramicroscopic particles dispersed through other substances such as emulsions and gels, which do not settle or cannot be separated by centrifuging as suspended solids (Jenkins, 1998; Ramprasad and Philip, 2016). While particles are solid materials which could be food, sand, soil, fibres, hair, and zeolites present in greywater from kitchen sink, washing machine, and laundry discharges.

A significantly high turbidity has been noticed in laundry wastewater at the washing stage, 39–296 NTU, compared with the rinsing stage, 14–29 NTU. In general, the turbidity of greywater from mixed resources was reported within 15.3–240 NTU (Christova–Boal et al., 1996). While the highest values of suspended solids have been reported from laundry and kitchen wastewaters varying in concentration within the range

17–330 mg/l, compared with 120–450 mg/l for general domestic wastewater (Henze et al., 2001).

Table 2.2. Greywater sources and associated contaminants (WHO, 2006b).

Greywater Source	Possible Contents
Automatic clothes washer	Suspended solids (dirt, lint), organic material, oil and grease, sodium, nitrates and phosphates (from detergent), increased salinity and pH, bleach
Automatic dishwasher	Organic material and suspended solids (from food), bacteria, increased salinity and pH, fat, oil and grease, detergent
Bathtub and shower	Bacteria, hair, organic material and suspended solids (skin, particles, lint), oil and grease, soap and detergent residue
Sinks, including kitchen	Bacteria, organic matter and suspended solids (food particles), fat, oil and grease, soap and detergent residue

Despite the turbidity and suspended solids values of greywater is lower than in municipal wastewater, clogging risk of recycling and/or treatment systems could be caused by colloids and particles (Mays and Hunt, 2005). Because surfactants in detergents prevent colloids to agglomerate due to sorption of the surfactants on the surface of the colloids producing solids in stabilised phase (Myers, 1988; Garland et al., 2004).

2.2.2. Chemical Properties

The quality of supplied water has a significant effect on the chemical properties of domestic grey wastewater, (Tables 2.1–2.3). The pH and alkalinity in greywater generated from laundry have been indicated to be relatively higher than from other sources (Friedler and Galil, 2003), with a range value between 9.3–10.0 (Christova–Boal et al., 1996; F. Li et al., 2009; Houshia et al., 2012), while for general raw greywater the range was 5.0–10 (Nghiem et al., 2006).

Although alkalinity and hardness are widely evaluated for drinking water, they also give vital indications of clogging problems, similarly to turbidity and suspended

solids contents, for system accessories when greywater is reused or treated (Jeppesen, 1996).

Infiltration of greywater on soil will change the soil properties, such as natural buffering and sorption capacity of pollutants. The alkalinity, hardness, and pH of the infiltration water could determine the resulting pH and buffering capacity of the soil (Delle Site, 2001). However, the influence of chemicals added during the use of the water is not limited in relation to those parameters.

Furthermore, the conventional wastewater quality measurements, such as 5–day biochemical oxygen demand (BOD_5), chemical oxygen demand (COD), and the nutrient concentrations (nitrogen and phosphorus) are also other indications for the risk of pollution when greywater is intended for reuse or treatment, depending on both resource of greywater and recycling purpose (Jefferson et al., 2004; Pidou et al., 2007).

Oxygen content in greywater can be estimated from BOD_5, COD, and dissolved oxygen (DO) as well. However, the COD is extracted from chemicals used for personal hygiene, washing dishes, and laundry detergents (Eriksson et al., 2002). Oxygen in wastewater is consumed by organism activities such as metabolism and respiration through degradation of organic matters during storing and transport producing thereby sulphide and leading to oxygen depletion (Brix et al., 2007; Winward et al., 2008).

The published measurements of 5–day biochemical oxygen demand and chemical oxygen demand are summarised in Table 2.1. It has been clearly noticed that there are wide differences in values for various greywater resources. BOD_5 ranges were between 5–300, 48–472, 536–1460, and 33–1460 mg/l, while, the ranges of COD were 100–633, 12.8–2950, 3.8–1380, and 3.8–2263 mg/l, for bathroom, laundry, kitchen, and mixed raw greywater, respectively.

Nitrogen as a nutrient in wastewater is usually determined in terms of ammonium–nitrogen (NH_4–N), nitrite–nitrogen (NO_2–N), and nitrate–nitrogen (NO_3–N). Although urine is the main source of nitrogen in domestic wastewater and should not be present in greywater, kitchen effluents contribute significantly to elevation of nitrogen content in greywater but it remains lower than nitrogen in combined wastewater (Jefferson et al., 2004; Kariuki et al., 2012). In general, a lower level of nitrogen has been reported from bathroom and laundry, 0.1–15.0 and 0.04–11.3 mg/l for NH_4–N, respectively, and for values of NO_3–N the ranges were between 0.05–6.3 and 0.4–7.5 mg/l, respectively. While, the reported NH_4–N and NO_3–N for kitchen discharge were in the range between 0.002–23.0 and 0.3–5.8 mg/l, respectively. The corresponding values for mixed resources of greywater were 0.03–25.4 and 0.0–38.0 mg/l, respectively, (Table 2.1).

Phosphorus is a reactive element and is never found in its elemental form (Emsley and Hall, 1976), however, phosphorus only exists in its pentavalent forms, such as orthophosphate and polyphosphate, in the aquatic ecosystem (Correll, 1998). For wastewater, washing discharges with different types of detergents are the primary sources of phosphorus in greywater (Jeppesen, 1996), however, degradation of vegetables and other food partials could result in phosphorus in kitchen greywater. The observed concentrations of total phosphorus measurements for greywater were between 4–14 mg/l caused by washing detergents, (Table 2.2). While the corresponding range for traditional wastewater was 6–23 mg of total–P per litre. In Table 2.1, the reported ortho–phosphate–phosphorus (PO_4–P) ranges were 0.94–48.8, 4.0–171.0, 12.7–32.0, and 0.02–68.0 mg/l for bathroom, laundry, kitchen, and mixed resources of greywater (Eriksson et al., 2002; Ramona et al., 2004).

2.2.3. Anticipated Chemicals in Greywater

There is a wide range of chemical composition products used for domestic purposes which have negative effects on the environment since they are released into wastewater (Daughton and Ternes, 1999).

Greywater generated from kitchens is always contaminated with lipids (fat, oils, and steroids), dairy products, coffee, tea, glucose, and washing products, while laundry greywater contains different types of detergents, soaps, shampoos, perfumes, bleaches, preservatives, waxes, dyes and cleaners. As a result, a heterogeneous group of compounds will be expected to appear in greywater constitutes, known as xenobiotic organic compounds (XOCs), (Eriksson, 2002; Press–Kristensen, et al., 2007).

Characteristics of greywater could be estimated from the information pertaining to ingredients which is available in the declaration of contents on different types of common household chemical products (Eriksson et al., 2003).

Among different groups of chemicals, surfactants used in detergents are indicated as a major combination, which include non–ionic, anionic and amphoteric surfactants, since cleaning products are expected to be consumed in large quantities worldwide (Levine et al., 2005). Degradation processes in greywater by oxidation and microorganisms may introduce other compounds differing from the original ones, in addition chemicals reacting with each other may result in new compositions (Dixon et al., 2000).

Food products may also contain anther large group of chemical combinations such as fragrances and flavours, solvents and preservatives (Eriksson et al., 2002, 2003). The preservatives are biocide fungicides used in a wide range of manufactured food and are toxic in some concentrations (Kahle et al., 2008).

Table 2.3. Trace element concentrations (mg/l) of real greywater.

Reference	Greywater Source	Aluminium	Boron	Calcium	Potassium	Magnesium	Sodium	Sulphur	Silicon	Phosphorus
Eriksson et al. (2002)	Bathroom GW	≤0.1	≤0.1	3.5–7.9	1.5–5.2	1.4–2.3	7.4–18.0	1.2–3.3	3.2–4.1	0.11–1.2
	Laundry GW	≤1.0–21	0.1–0.5	3.9–14.0	1.1–17.0	1.1–3.1	44–480	9.5–40.0	3.8–49.0	0.062–57.00
	Kitchen GW	0.67–1.8		13–30	19–59	3.3–7.3	29–180			0.06–74.0
	Mixed GW	0.10–3.55		11–35	6.6	1.5–19.0	21–230			0.16–27.3
Christova–Boal et al. (1996)	Bathroom GW	≤1.0		3.5–7.9	1.5–5.2	1.4–2.3	7.4–18.0	1.2–3.3	3.2–4.1	0.11–1.80
	Laundry GW	≤1.0–21.0		3.9–12.0	1.1–17.0	1.1–2.9	49–480	9.5–40.0	3.8–49.0	0.062–42.0
F. Li et al. (2009)	Bathroom GW	2.44		33.8	8.1	5.74		23.7		
	Laundry GW	0.49		60.79	11.2–23.3	6.15		19		
	Kitchen GW	0.003		47.9	5.79	5.29		16.3		
Ramona et al. (2004)	Shower GW	0.03	0.14	71.0–93.6	9.8–12.4	43.2–50.0	93.0–142.7			
Nghiem et al. (2006)	Real GW			3.6–200.0						
Houshia et al. (2012)	Raw GW			89.5	37.3	132.2				
Leal et al. (2012)	Raw GW			42.8	14.5	11.6	128			
Kariuki et al. (2012)	Kitchen GW1			4.9	23.4	4.8	15.38			
	Laundry GW1			1.3	26.9	2.54	39.23			
	Bath GW2			0.96	10	0.27	6.15			
	Kitchen GW2			0.93	16.9	0.28	9.89			
	Laundry GW2			0.32	31.8	1.14	35.38			
Jefferson et al. (2001)	Real GW	0.003		47.9	5.79	5.29		16.3		

2.2.4. Heavy Metals and Other Trace Elements

Among the other pollutants, the content of heavy metals (e.g. Al, Fe, Mn, Cd, Cu, Pb, Hg, Zn, Ni, and Cr) and XOCs will be of importance. The concentration of metals and other elements will largely be dependent on the concentrations and quality of the water from the water works. Laundry wastewater has been found to contain elevated sodium levels compared to other types of grey wastewater. The sodium in the laundry wastewater may be caused by the use of sodium as a counter ion to several anionic surfactants used in powder laundry detergent (Jeppesen, 1996), and the use of sodium chloride in ion–exchangers. Only relatively low amounts of heavy metals have been reported in the literature, with one exception being Christova–Boal et al. (1996) who found notably high levels of zinc in grey wastewater. The laundry wastewaters contained 0.09–0.34 mg/1 while the bathroom wastewater contained 0.2–6.3 mg Zn/l, (Table 2.3). The other authors found concentrations in the range < 0.01–1.8 mg/l. One reason for the high values in the bathroom wastewater could be some chlorine tablets that had been used for disinfecting. These tablets are acidic and that may cause leaching of zinc from the plumbing. Heavy metals are linked to personal care and cosmetic products (Bocca et al., 2014). Furthermore, human hair has been assessed for its content in terms of metals as a function of different washing products (Chojnacka et al., 2012). Varying concentrations of metals can be linked to discharges of baths, showers and hand washing basins, while kitchen discharge is commonly less rich in metals (Kariuki et al., 2012). In general, P, Na, Fe, Cu, Cd and Cr concentrations were highest in laundry greywater. The highest values of Zn and Mg were found in kitchen greywater from households without sewers (Kariuki et al., 2012). Other unexpected sources of metals are linked to pharmaceutical products and rechargeable batteries.

Table 2.4. Heavy metal concentrations (mg/l) of real greywater.

Reference	Greywater source	Cadmium	Chromium	Copper	Iron	Manganese	Nickel	Lead	Zinc	Molybdenum
Eriksson et al. (2002)	Bathroom GW	0.00054–0.010		0.06–0.12	0.34–1.10			0.003	0.06–6.3	
	Laundry GW	0.00036–0.038	≤0.025	≤0.050–0.322	0.29–1.00	0.029	≤0.028	0.033 to ≤0.063	0.09–0.44	
	Kitchen GW	0.00052–0.007	≤0.025–0.130	0.05–0.26	0.6–1.2	0.031–0.075	≤0.025	0.005–0.140	0.096–1.8	
	Mixed GW	≤0.006–0.030	≤0.01026–0.05	0.018–0.230	<0.05–4.37	0.014–0.075	≤0.015–0.050	≤0.01–0.15	≤0.01–1.6	
Christova–Boal et al. (1996)	Bathroom GW	≤0.01		0.06–0.12	0.34–1.10				0.2–6.3	
	Laundry GW	≤0.01		≤0.05–0.27	0.29–1.00				0.09–0.32	
F. Li et al. (2009)	Bathroom GW			0.0618	0.36	0.0121			0.0644	
	Laundry GW			0.08	0.11	≤0.05			0.00	≤0.05
	Kitchen GW			0.006	0.017	0.04			0.03	0.00
Al–Hamaiedeh and Bino (2010)	GW Range							1.00–1.31		
	GW Average	0.008						1.19		
Ramona et al. (2004)	Shower GW 1	≤0.02	≤0.02	≤0.02	0.19	≤0.02	≤0.02	≤0.02	0.18	≤0.02
	Shower GW2	≤0.02	≤0.02	≤0.02	0.06	≤0.02	≤0.02	≤0.02	0.03	≤0.02
Eriksson et al. (2010)	Raw GW1	0.0001		0.0087–0.0110			0.007–0.039	0.0025–0.0031		
	Raw GW2	≤ 0.0001–0.0090		0.0085–0.0250			0.0055–0.0079	0.0018–0.0032		
Leal et al. (2012)	Raw GW			0.0906	0.29			≤0.010		
Kariuki et al. (2012)	Kitchen GW1	5.5	16.1	0.9	1.9	1.4		0.9	6.6	
	Laundry GW1	7	0.9	1	3.6	0.4		0.8	0.4	
	Bath GW2	10.7	11.1	2.6	3.8	0.3		0.2	0.2	
	Kitchen GW2	10	11.3	2.3	9.7	0.2		0.3	0.1	
	Laundry GW2	11.2	16.1	2.9	17.5	0.3		0.0	0.7	

2.2.5. Microorganisms

The risk of presence, diffusion, and survival of microorganisms (pathogenic viruses, bacteria, protozoa and helminths) in greywater is of great concern due to their relevant health and environmental impacts (Vasconcelos and Swartz, 1976). Microorganisms may be introduced in the greywater system either during usage or from regrowth (Ottoson and Stenström, 2003). Although toilet effluent is excluded, washing hands after toilet use, and washing and changing nappies, may contribute to the presence of excreta in greywater (Benami et al., 2016). Furthermore, Eriksson et al. (2002) reported that microorganisms may be passed through elderly people suffering from susceptible infection or the human immunodeficiency virus (HIV–positive). Contact with animals or sources of infection, such as persons with a particular health disease can spread and transfer other pathogens to greywater. *Escherichia coli* content is one of several indicators of faecal pollution in greywater as well its associated viruses (Maimon et al., 2010). Main sources of microorganisms in kitchen wastewater are raw meat and contaminated uncooked meals, where *E. coli* contents have been evaluated in the range 1.3×10^{5}–2.5×10^{8} per 100 ml, *thermotolerant coli* in the range 9.4×10^{4}–3.8×10^{8} per 100 ml, and faecal streptococci between 5.1×10^{3} and 5.5×10^{8} per 100 ml. The microorganism amounts were found to be slightly lower in bathroom greywater than kitchen greywater, with values of 3.0×10^{3} per 100 ml *faecal coliforms*, 70–2.4×10^{7} per 100 ml of total coliforms, and 1–7×10^{4} per 100 ml of *faecal streptococci*. While laundry wastewater was found to contain 9×10^{4}–1.6×10^{4} per 100 ml faecal coliforms, 8.9×10^{5} per 100 ml of total coliforms, and *faecal streptococci* in the range 1–1.3×10^{6} per 100 ml (Eriksson et al., 2002).

Many species of water and excreta pathogens associated with microorganisms have been indicated in different types of water including greywater.

Occasionally, traces of urine are present in bathroom greywater with the possibility to transfer infections and pathogens through urinary *schistosomiasis* (Schistosoma haematobium), *typhoid* (Salmonella typhi) and *leptospirosis* (Leptospira), which are bacteria (Feachem, 1980).

Candida albicans, *Pseudomonas aeruginosa* and *Staphylococcus aureus* have been observed in wastewater generated from showers, because these microorganisms are commonly found in the human mouth, nose and throat (Burrows et al., 1991). Furthermore, *Campylobacter spp, Cryptosporidia*, *Giardia,* and *Salmonella spp* (Christova–Boal et al., 1996), as well as *Shigella* and *Entamoeba histolytica* have been indicated in shower greywater (Sheikh, 1993).

Parasites such as roundworms and other viruses (*Hepatitis* and *enteroviruses*) could appear with host bacteria like *Salmonella typhi* and *Salmonella paratyphi* (WHO, 2006a).

The majority of diffusion of microorganisms to the environment is either from disposal or reuse of wastewater. If greywater is used for surface irrigation and toilet flushing, Legionella is considered as a serious threat to human health through inhalation, since it is spread by aerosols.

These helminths can infect humans but they cannot proliferate within the host, with the exception of *Strongyloides* (Feachem, 1980). Parasitic protozoa and helminths will not be a problem if greywater is reused for subsurface irrigation or infiltration. Due to their large size, parasitic protozoa and helminths will be removed by filtration as the water percolates under gravity (Dixon et al., 1999). *Cryptosporidium* and *Giardia*

(protozoa) can survive and are relatively resistant to densification, however, *Clostridium perfringens* (protozoa) spreads and lives longer than other organisms through the germs which are indicators of faecal contamination (Feachem, 1980).

Through a study investigating the survival of *Salmonella typhimurium*, *Shigella dysenteriae* and poliovirus in greywater, it was indicated that the numbers of *Salmonella* and poliovirus did not change for two days and subsequently decreased, while, the numbers of *Shigella* decreased more rapidly (Rose et al., 1991).

2.3. Synthetic Greywater

It is well known that a single source might be very misleading since the concentration varies over the day and is different on different days of the week (Eriksson et al., 2002). It is highly important to evaluate the data in the literature, (Tables 2.1–2.3), in order to get some typical values or ranges for each parameter in the different types of grey wastewater. That kind of information would be appreciated and useful in evaluation of the best method for treatment or in a risk assessment of reuse of grey wastewater.

Unfortunately, the information available is still too limited for most of the parameters. The increased focus on the treatment and reuse of highly variable real greywater has driven some researchers to create greywater with stable properties artificially (Hourlier et al., 2010) as indicated in Table 2.4.

Some synthetic greywaters have been created by mixing different recipes of chemical products that households use and/or analytical grade chemicals known to be present in real greywater. Consequently, these chemicals are expected to control the characteristics of the generated greywater in terms of water quality (Schäfer et al., 2006). The pathogens and nutrients occurring in greywater are present in much lower concentrations than in blackwater (Eriksson et al., 2002).

In fact, greywater does not contain the right nutrient and trace element ratio required for standard biological treatment or advanced treatment by membrane bioreactor (Al–Jayyousi, 2003; F. Li et al., 2009; Jefferson et al., 2001). Furthermore, low concentrations of trace elements have been linked to greywater (Eriksson et al., 2002).

Nghiem et al. (2006) investigated the feasibility of submerged ultrafiltration technology applied for greywater recycling. The synthetic greywater solution contained kaolin, cellulose, humic acid, sodium hypochlorite, calcium chloride electrolyte and a sodium bicarbonate buffer. These materials were also used in combination with sodium dodecyl sulphate to represent the synthetic greywater proposed by Schäfer et al. (2006).

Nazim and Meera (2013) studied the treatment ability of a synthetic greywater by adding different concentrations of an enzyme protein solution to examine the reduction of chemical variables including nutrients. The mixture of synthetic greywater contained glucose, sodium acetate trihydrate, ammonium chloride, disodium hydrogen phosphate, potassium dihydrogen phosphate, magnesium sulphate, and cow dung.

Diaper et al. (2008) introduced a synthetic greywater recipe to simulate combined laundry and bathroom greywater from an Australian residential dwelling. The constituents of the greywater included a variety of personal hygiene and household products, some laboratory grade chemicals (sodium dodecyl sulphate, sodium hydro carbonate, sodium phosphate, boric acid, and lactic acid), and secondary sewage effluent sourced from a local wastewater treatment plant.

Fenner and Komvuschara (2005) described a new approach to model the effect of factors influencing ultraviolet disinfection efficiency of real and synthetic greywaters. A range of synthetic greywater recipes was developed for both soft and hard waters to ensure they were representative of the properties of real greywater samples. A typical

synthetic greywater recipe comprised dextrin, ammonia chloride (NH_3Cl), yeast extract, soluble starch, sodium carbonate (Na_2CO_3), monosodium phosphate (NaH_2PO_4), potassium phosphate (K_2PO_4) and an *Escherichia coli* culture mixed with distilled water.

Surendran and Wheatley (1998) proposed a biological treatment process for greywater obtained from large buildings. The synthetic greywater used comprised a known amount of soap, detergent, starch yeast extract and cooking oil. Settled sewage was also added to provide appropriate bacteria counts.

Jefferson et al. (2001) dosed synthetic and real greywater with nutrient supplements. The synthetic greywater recipe comprised synthetic soap, hair shampoo, sunflower oil and tertiary effluent.

Gross et al. (2006) developed a new small–scale vertical–flow constructed wetland for decentralised treatment of greywater. The removal of indicator and pathogenic microorganisms was investigated to assess the reuse of treated greywater for irrigation purposes. The focus was on the removal dynamics of *Escherichia coli*, *Staphylococcus aureus* and *Pseudomonas aeruginosa* in three different synthetic greywaters. Each greywater was made by combining three waste stocks representing laundry, bath and kitchen wastes. The composition of synthetic greywater for each stock contained laundry soap, shampoo, cooking oil, and kitchen effluent (comprising one egg and one tomato). All greywater types were supplemented with raw sink effluent from a large dining room. This effluent, which contained an inoculum of *E. coli* and other bacteria, was added in a small enough volume not to affect the composition of the synthetic greywater (Gross et al., 2007a).

In a controlled study, a recirculating vertical–flow constructed wetland was investigated to assess the effect of irrigation with treated greywater on soil properties

(Travis et al., 2010). The greywater was prepared according to a similar recipe used by Gross et al. (2007a). However, pulverised bar soap was applied instead of shampoo in the synthetic greywater.

Gross et al. (2007b) developed an economically sound, low–tech and easily maintainable combined vertical–flow constructed wetland and trickling filter system for greywater treatment and subsequent recycling. The greywater was prepared artificially by mixing laundry detergent, boric acid and raw kitchen effluents into tap water.

Comino et al. (2013) proposed a functional hybrid phytoremediation pilot platform for the treatment of greywater. The pilot plant was tested with and without vegetation for different design specifications as well as for various organic and hydraulic loads of synthetic greywater. This study by Comino et al. (2013) followed one by Gross et al. (2007b) in terms of preparation of artificial greywater.

Glasshouse experiments were conducted by Pinto et al. (2010) to understand the effects of greywater reuse for irrigation of plants. Changes in soil pH, electrical conductivity and nutrient content (total nitrogen and total phosphorus) due to greywater irrigation were assessed. Synthetic greywater was prepared by mixing a commonly available local detergent with potable water.

Winward et al. (2008) evaluated three treatment technologies, constructed wetlands, membrane bioreactors and membrane chemical reactors, for indicator microbial removal and greywater reuse potential under conditions of low and high strength greywater influents. A high strength supplementary solution together with real greywater was pumped to the treatment systems. Real greywater was referred to as low or high strength solution based on a mixture of locally sourced shampoo diluted by tap water.

2.3.1. Chemicals Used in Greywater Simulation

The concentrations of the corresponding greywater pollutants (e.g., organic strength, nitrogen, phosphorus, surfactants and metals) as a result of mixing the ingredients listed have been published in the references shown in Tables 2.5 and 2.6 showing their corresponding water quality. However, most recipes cannot be reproduced accurately, because the environmental boundary conditions are variable or unreported.

Moreover, some ingredients, such as cow dung, shampoo and kitchen effluent, are unspecified. A reproduction of the published water quality data is therefore of little use to the researchers of the introduced formulas. Nevertheless, a review of the most common chemicals used for artificial greywater recipes is summarised below.

Kaolin is a common clay mineral composed of alternating sheets of aluminium hydroxide and silicate (Essington, 2004). It is frequently selected as an artificial greywater component to represent suspended organic and inorganic solids in greywater, which may originate from natural clay containing various mineral components. These solids are often generated from kitchen and laundry effluents (Eriksson et al., 2002). Kaolin is also used in synthetic wastewater recipes (Nghiem et al., 2006; Schäfer et al., 2006; Marfil–Vega et al., 2010; Fitria et al., 2014).

Cellulose is the principal structural component of plant cells and leaves. Furthermore, the majority of the carbohydrates found in soils are derived from cellulose, which is one of many polymers found in nature (Essington, 2004). Cellulose is frequently chosen to mimic organic fibres in greywater, since kitchen sinks and dishwashers are common sources of organic fibres (Nghiem et al., 2006; Schäfer et al., 2006).

All natural waters contain humic (Essington, 2004) constituents as the result of biodegradation of animal and plant matter or they might form in situ due to the presence

of soils, nutrients, and cellulosic substrates for microbial action in the waste (Wall and Choppin, 2003). Humic acid is often used to represent dissolved organic matter in greywater (Nghiem et al., 2006; Schäfer et al., 2006).

Boric acid is frequently applied to represent boron ions in greywater. One source of boron is natural and the other is a result of human activities (e.g., extraction plant, industry and detergent containing sodium perborate). It follows that many water sources and wastewaters may contain boron in variable concentrations (Gross et al., 2007b; Diaper et al., 2008).

The following salts have been previously suggested as possible ingredients in synthetic greywater: Sodium chloride (dissolved monovalent salt) is found as a common ingredient of soap solutions and dyes (WilliamáMcBain et al., 1912; Myers, 1988). Sodium hydrogen carbonate (natural buffer) and sodium dodecyl sulphate are mainly used for the manufacture of detergents. Their greatest cleaning application is as a filler in powdered home laundry detergents (Myers, 1988; Zhu et al., 2015).

Sodium hydrogen carbonate, sodium dodecyl sulphate and sodium phosphate are important in the manufacture of textiles by reducing negative charges on fibres, so that dyes can penetrate evenly (Syafalni et al., 2012). Some of these salts have previously been used in synthetic grey and municipal wastewater recipes (Surendran and Wheatley, 1998; Nghiem et al., 2006; Schäfer et al., 2006; Diaper et al., 2008; Fenner and Komvuschara, 2005; Fitria et al., 2014).

Table 2.5. Recipes reported for different synthetic greywaters.

Reference	**Surendran and Wheatley (1998)**		**Diaper et al. (2008)**		**Nazim and Meera (2013)**		**Fenner and Komvuschara (2005)**	
Country	UK		Australia		India		UK	
Treatment approach	Multi–stage bio–filter		– Biological with suspended media – Chemical flocculants, ultraviolet disinfection and filtration – Settling, biological with fixed media		Using garbage enzyme after filtration		Ultraviolet disinfection system	
Dextrin	85 mg/l		Sunscreen or moisturiser	15 or 10 mg/l	Glucose	300 mg/l	Dextrin	85 mg/l
Ammonium chloride	75 mg/l		Toothpaste	32.5 mg/l	Sodium acetate trihydrate	400 mg/l	Ammonium chloride	75 mg/l
Yeast extract	70 mg/l		Deodorant	10 mg/l	Ammonium chloride	225 mg/l	Yeast extract	70 mg/l
Soluble starch	55 mg/l		Sodium sulphate	35 mg/l	Sodium dihydrogen phosphate	150 mg/l	Soluble starch	55 mg/l
Sodium carbonate	55 mg/l		Sodium hydrogen carbonate	25 mg/l	Potassium dihydrogen phosphate	75 mg/l	Sodium carbonate	55 mg/l
Washing powder	30 mg/l		Sodium phosphate	39 mg/l	Magnesium sulphate	50 mg/l	Sodium dihydrogen phosphate	11.5 mg/l
Sodium dihydrogen phosphate	11.5 mg/l		Clay (Unimin)	50 mg/l	Cow dung	225 ml/l	Potassium phosphate	4.5 mg/l
Potassium sulphate	4.5 mg/l		Vegetable oil	0.7 mg/l			*Escherichia coli* culture	15 ml/l
Settled sewage	10 ml/l		Shampoo/hand wash	720 mg/l				
Shampoo	0.1 ml/l		Laundry	150 mg/l				
Cooking oil	0.1 ml/l		Boric acid	1.4 mg/l				
Biochemical oxygen demand	approx. 200 mg/l		Lactic acid	28 mg/l				
			Secondary effluent	20 ml/l				
Reference	**Gross et al. (2007) / Comino et al. (2013)**		**Nghiem et al. (2006)**		**Jefferson et al. (2001)**		**Hourlier et al. (2010)**	
Country	Israel/Italy		Australia		UK		France	
Treatment Approach	Vertical–flow constructed wetland/ Hybrid constructed wetland		Submerged ultrafiltration membranes		Membrane bioreactors and activated sludge systems		Direct membrane nano–filtration	
Laundry detergent	20 g		Humic Acid	20 mg/l	Synthetic soap	0.64g	Lactic acid	100 mg/l
Boric acid	0.86 g		Kaolin	50 mg/l	Hair shampoo	8.0 ml	Cellulose	100 mg/l
Kitchen effluent	400 ml		Cellulose	50 mg/l	Sunflower oil	0.1 ml	Sodium dodecyl sulphate	50 mg/l
Tap water	150 l		Calcium chloride	0.5 mM	Tertiary effluent	24 ml	Glycerol	200 mg/l
			Sodium chloride	10 mM	Tap water	10 l	Sodium hydrogen carbonate	70 mg/l
			Sodium hydrogen carbonate	1 mM			Sodium sulphate	50 mg/l
							Septic effluent	10 mg/l

Calcium nitrate and calcium chloride have been suggested as components in synthetic greywater. Calcium salts are chosen to provide calcium ions to artificial greywater. Previous research used calcium salts in synthetic greywater (Nghiem et al., 2006; Schäfer et al., 2006).

Laboratory grade chemicals such as potassium nitrate, mono–potassium phosphate and magnesium sulphate have been chosen in previous studies (Fenner and Komvuschara, 2005; Nazim and Meera, 2013) to resemble real greywater in terms of nutrients and macronutrients generated from laundry and kitchen effluents.

Low suspended solids and turbidity linked to greywater indicates that a large proportion of pollutants are dissolved. Although organics present in artificial greywater are relatively similar to domestic greywater, their chemical natures are quite different. The deficiency of nutrients and low values of biodegradable organic matter limit the effectiveness of biological treatment of greywater (Al–Jayyousi, 2003).

Iron(III) chloride, manganese(II) chloride, chromium(III) nitrate, zinc sulphate, copper sulphate, cadmium oxide, nickel oxide, and lead(II) oxide are commonly selected to provide heavy metals to artificial greywater, as discussed in publications reported in Table 2.3.

Sources of heavy metals in real greywater may be from cosmetics (Eriksson et al., 2010), other products such as skin emulsions (creams, lotion and jelly), soap, shampoo, hair cream, henna dye (Chauhan et al., 2010; Bocca et al., 2014), and from body parts such as hair, nails and dead skin cells (Eriksson et al., 2002; Chojnacka et al., 2012).

Ammonium molybdate tetrahydrate is used to provide molybdenum in artificial greywater. Molybdate is also known to enhance the biological treatment of wastewater

(Jefferson et al., 2001). Sodium hydroxide and hydrochloride acid are widely used as buffers to adjust the pH value of a chemical solution.

Small quantities of secondary or tertiary effluent obtained from predominantly domestic wastewater treatment plants is frequently recommended as an additive to synthetic greywater to provide a source of pathogens and microorganisms in general (Diaper et al., 2008; Gross et al., 2007a; Hourlier et al., 2010; Fenner and Komvuschara, 2005; Fitria et al., 2014).

However, the addition of microbes might not be necessary for experiments in non–sterile environments such as outdoor trials where a microbial population adjusted to the system tested will establish naturally eventually.

One target of this study is to evaluate the stability of chemical compositions of artificial greywater through specific storage time experiments, without the contribution of biological treatment, which is offered by microorganisms.

There are numerous papers in the peer–reviewed literature indicating greywater recipes that have no artificially introduced microorganisms in the list of ingredients (Nghiem et al., 2006; Schäfer et al., 2006).

Table 2.6. Characteristics of published synthetic greywaters.

Parameter	Unit	Surendran and Wheatley (1998)	Diaper et al. (2008)	Nazim and Meera (2013)	Gross et al. (2007)	Comino et al. (2013)	Nghiem et al. (2006)	Hourlier et al. (2010)
Biochemical oxygen demand	mg/l	215	146.7	192	28.0–688			58–75
Chemical oxygen demand	mg/l		276.7	290	702–984	77.4		391–505
Ammonia–nitrogen	mg/l	11		9.6	0.1–0.5			
Nitrate–nitrogen	mg/l		<0.2		0.0–5.8			
Nitrite–nitrogen	mg/l		<0.003		0.0–1.0			
Total nitrogen	mg/l				25.0–45.2			
Ortho–phosphate–phosphorus	mg/l	4.9		110				
Total phosphorus	mg/l		17.8		17.2–27.0			
pH	–		7.4	6.16	6.3–7.0	7.3	7.5–8.0	6.29–7.29
Redox potential	mV							
Turbidity	NTU	72	52.1				140	4–42
Total dissolved solids	mg/l	12.3		563		247.4		
Total suspended solids	mg/l	196	59		85–285			41–87
Total organic carbon	mg/l	81.8	62.2					
Dissolved organic carbon	mg/l							106–149
Electrical conductivity	µs/cm		322.2		1000–1300	495.1		159–212
Dissolved oxygen	mg/l							
Aluminium	mg/l		1.6					
Boron	mg/l				1.4–1.7			
Calcium	mg/l		7.6					
Magnesium	mg/l		1.3					
Sodium	mg/l		65.3					
Surfactants	mg/l				4.7–15.6			33.5–69.8
Salinity	–					0.1		

2.4. Wetlands

2.4.1. Types of Wetlands

Constructed treatment wetlands are artificial manmade systems for remediation of different pollutants from wastewater through physical, chemical, and biological mechanisms, which are similar to those processes which occur in natural wetlands (Tchobanoglous, 1993; Scholz, 2006; Vymazal, 2007)

Natural wetlands, such as marshes, have been employed as appropriate approaches for disposal of collected wastewater and sewage since the 1900s (Brix, 1994), to become sources of accumulation and diffusion of nutrients and other contamination (Kadlec and Knight, 1996).

According to Vymazal (2005), the majority of research on wastewater treatment in wetlands vegetated with different macrophytes was carried out in Germany by Kathe Seidel in 1952. Additionally, in the early 1960s, the role of wetland macrophytes, different wastewater types, different sludge resources, and development approaches were explored in the treatment of unsatisfied systems such as ponds and septic tanks for rural and decontrolled wastewaters. Through the early system of the hydro–botanical method with the concept of Free Surface Flow (FSF) of horizontal and vertical constructed wetlands (Seidel, 1976), the wetland substrate bed of sludge has been improved with high hydraulic conductivity sandy soil vegetated with *P. australis* to introduce Sub–Surface Flow Constructed Wetlands (SSF–CWs), with either horizontal (HF–CWs) or vertical (VF–CWs) subsurface flow (Cooper et al., 1996; Vymazal, 2010). This improvement led consequently, with development of the Root Zone Method (RZM) by Reinhold Kickuth (Kickuth, 1977), to derive the basic concept for hybrid constructed wetlands (Brix, 1987). The first full–scale applications of FSF–CWs were established in the Netherlands in 1967

(Vymazal, 2010). However, FSF–CWs have not gained wide popularity in Europe where SSF–CWs dominated in the 1980s and 1990s (Vymazal and Kröpfelová, 2011). The arrangement of various types of constructed wetlands has reflected the combined manner of hybrid wetlands to achieve highly efficient treatment of wastewater (Vymazal, 2013a). Hence, the hybrid systems have prevailed as the multistage treatment of the early twenty–first century (Vymazal, 2005), as shown in Figure 2.3.

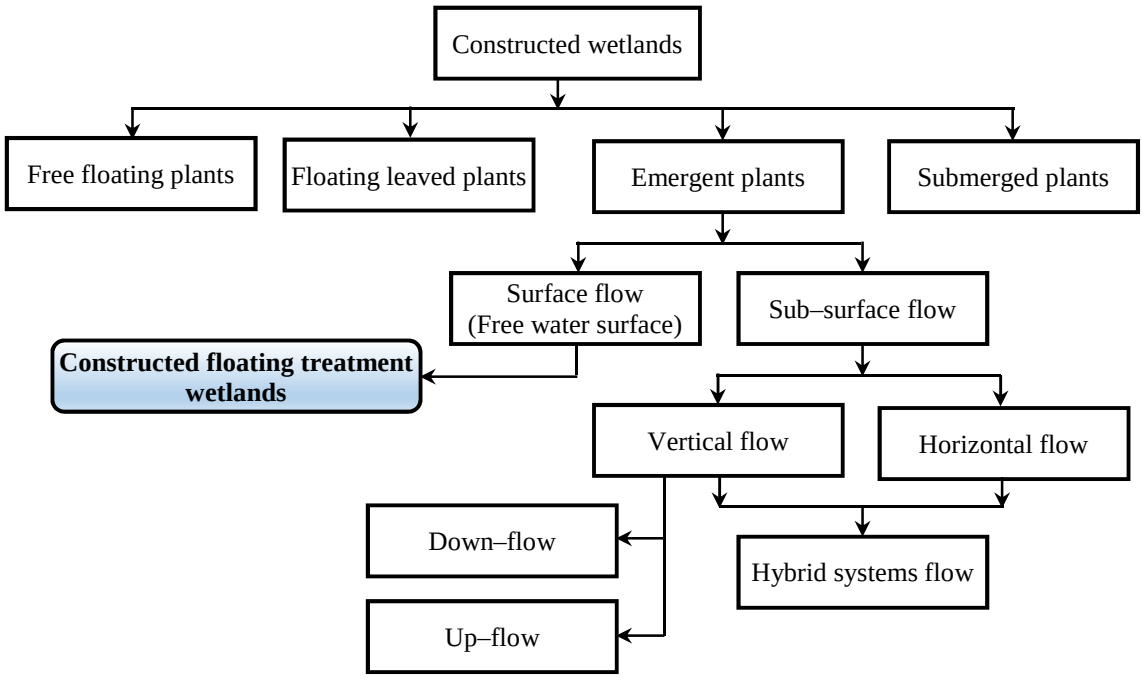


Figure 2.3. Classification of constructed wetlands for wastewater treatment (Vymazal, 2007).

Contamination and pathogens have been considered as serious threats to the environment, wildlife and water sources, in particular if they accumulate within the water bodies such as rivers and lakes. Runoff from streets, agricultural lands, and wastewater which may originate from industrial, commercial, or residential sectors can all be causes of pollutant diffusions that destroy the freshwater sources (Abdel, 1996; Campbell et al., 2004; Tota–Maharaj and Scholz, 2010). Among natural treatment systems such as wetlands, detention ponds have been used to mitigate the effects of various pollutants,

from stormwater in particular (Scholz, 2006; Aryal et al., 2010). From constructed wetland concepts, the role of macrophytes in the reclamation of wastewater has involved to produce artificial floating treatment wetlands to enhance aquatic life habitats (Nakamura and Mueller, 2008), but there has not been a comprehensive evaluation of their capacity for water quality improvement, despite these systems being in use from the early 1900s till the present day (Wang et al., 2015; Ladislas et al., 2015; Saeed et al., 2016).

2.4.2. Floating Treatment Wetlands (FTWs)

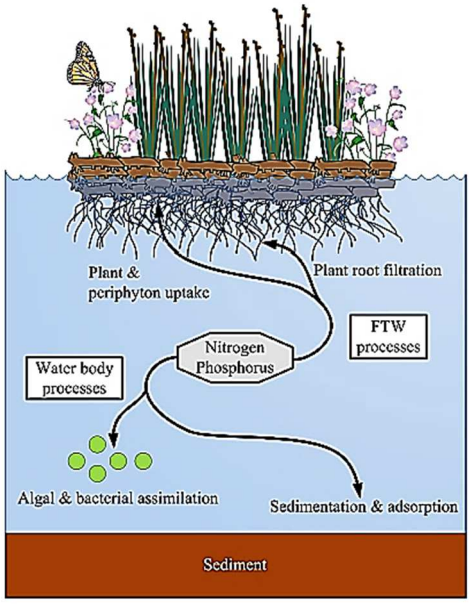


Figure 2.4. Schematic of an integrated FTW. The phosphorus and nitrogen are removed separately by the FTW and applied water body, such as river or retention pond (Wang and Sample, 2013).

Floating treatment wetlands (FTWs) are artificial floating mats, on the water surface of a pond, cultivated with aquatic vegetation in a hydroponic manner to simulate natural floating reedbeds (Kadlec and Wallace, 2009). The macrophyte roots are immersed within the water column without anchorage to the wetland substrate to provide a large surface area for microorganisms and the attached biofilm (Figure 2.4). Both roots

and microorganisms consume available organic and inorganic matters from polluted wastewater growth and metabolism processes (Brix, 1997).

Floating treatment wetlands are much like conventional constructed wetlands in terms of their practices and removal processes for treatment of various wastewater types such as municipal, industrial, agricultural, and stormwater (Hubbard et al., 2004; Billore et al., 2008; Duncan, 2009; Faulwetter et al., 2011). However, the floating vegetated mats in the former systems can be made of natural or industrial materials such as sludge, bamboo, polyethylene, or plastic (Hu et al., 2010; Van de Moortel et al., 2010; De Stefani et al., 2011; Tanner and Headley, 2011). There are some benefits which can be achieved in wastewater treatment with constructed floating wetlands, such as their low cost of construction, operation, and maintenance (Figure 2.5). There are no requirements for extra land, since the buoyant mat is suitable for fluctuating water level ponds (Faulwetter et al., 2011). Furthermore, using floating treatment wetlands gives an aesthetic improvement with the creation of a habitat for biodiversity and reduction of pollutants (Borne et al., 2013).

Figure 2.5. Artificial cultivated mat floated on pond water surface (Hartshorn et al., 2016).

Reducing eutrophication from surface waters and wastewater has directed consideration toward constructed wetlands for their natural mitigation of pollution by removing of wide variety of contamination (Scholz and Xu, 2002; Scholz et al., 2007; Carty et al., 2008). Organic compounds, suspended solids, pathogens, metals, and nutrients are common substances dominating different types of wastewater, in particular, according to the source of production, such as municipal, acid mine drainage, industrial, agricultural and stormwater runoff, effluent from livestock operations, as well as pharmaceutical and personal care products (Wei et al., 2008; Van de Moortel et al., 2010; Choudhary et al., 2011; Dunne et al., 2012; Teng et al., 2012).

However, these conventional wetlands are not likely to be applied in situations where land area is limited, such as in large cities, and land acquisition costs limit their broader use (Keizer–Vlek et al., 2014). A relatively new and evolving treatment practice may represent a significant opportunity to retrofit existing stormwater facilities by combining the functions of constructed wetlands and conventional retention ponds (Figure 2.5). This hybrid system is known as a floating treatment wetland (Tanner and Headley, 2011; Wang and Sample, 2014).

The first buoyant mat structure in natural floating wetlands consisted of colonies of crumbled biomass, roots of aquatic vegetation, and delamination of unvegetated, floating organic substrates from the deeper sediment reaching the surface of water in the form of islands. Then, sloughed plants due to flood or newly germinated seeds emerging, only occurs in a second phase, within those islands held together by rhizomes and stems to comprise floating reedbeds (Mallison et al., 2001; Adams et al., 2002; John et al., 2009).

The second type of formation occurs when the rhizomes of aquatic plants begin to colonise the water surface from a nucleus formed by floating aquatic vegetation that is either unattached or expanding from the shore line. The third formation type occurs when units of rooted vegetation and substrate split simultaneously from the bed, usually as a result of flooding as well (Somodi and Botta–Dukát, 2004).

In floating treatment wetlands, emergent macrophytes grow on a floating mat or buoyant frame that is placed on the water surface (Boutwell, 1995). In contrast to the more conventional wetland systems, the emergent macrophytes are not anchored in the sediment of the wetland, instead, their roots hang into the water column in a hydroponic manner (Figure 2.6).

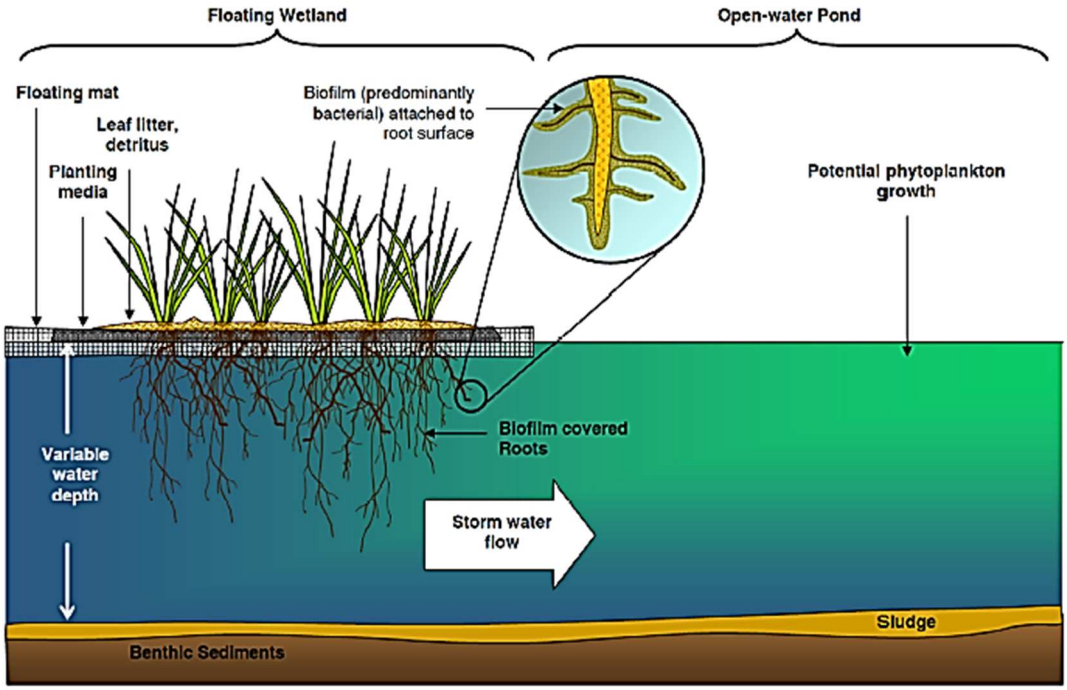


Figure 2.6. Schematic of an artificial floating island (also called integrated floating treatment wetland or integrated ecological floating bed (Yeh et al., 2015)).

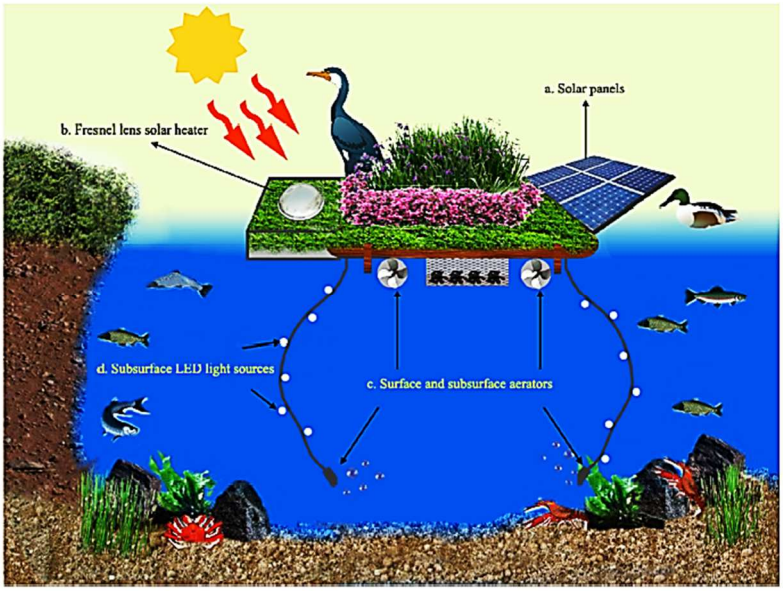
The entire underwater surface of the plants serves as a base for the attachment of microorganisms, which favours the breakdown of organic matters and the entrapment of suspended solids (X. Li et al., 2010; Keizer–Vlek et al., 2014). Phytoremediation using

floating macrophytes, floating–leaf macrophytes, emergent macrophytes, and, particularly, submersed macrophytes, is considered crucial to regulating the lake biological structure as macrophytes limit algal growth by competing for nutrients and sunlight and also increase herbivorous fish biomass by providing oxygen, food and refuge (X. Li et al., 2010; De Stefani et al., 2011).

Other advantages of floating treatment wetlands are their ability to cope with fluctuations in water levels, their aesthetic value (especially when using flowering plants), provision of habitat for invertebrates, fish and birds, and the sense of green they create in the city (Keizer–Vlek et al., 2014). In addition, the plants are forced to absorb nutrition directly from the water column, as they are not rooted in any substrate, which may improve the uptake rates of nutrients into biomass. Moreover, the by–product of plants can be easily harvested and subsequently used as food or be processed into biogas, bio–fertiliser, etc. (Hu et al., 2010).

Figure 2.7. Floating treatment wetlands for water and aesthetic improvement (Chang et al., 2013).

In conclusion, floating treatment wetlands hold a permanent pool of water and offer many beneficial uses including flood mitigation, pollution prevention, downstream erosion control, increased aesthetics, and recreational uses (Figure 2.7). Although the removal of nutrients is generally low for stormwater wet detention ponds in urban areas, floating treatment wetlands can be installed to offer an innovative solution (Figure 2.8) toward naturally removing excess nutrients and aiding in stormwater management (Hartshorn et al., 2016).



(a) solar panels to generate power for aerators and lights, (b) solar heaters to warm up water pockets for temperature sensitive species when needed, (c) surface and subsurface aerators to supply air for healthier oxidation, and (d) subsurface LED light source to adjust photo conditions for target species.

Figure 2.8. Floating treatment systems as enhancement of biodiversity with modification of schematic (Yeh et al., 2015).

Of course, there are certain disadvantages of using the planted floating–bed in lake restoration. First, it is difficult to control the hydraulic retention time and the pollutant loading rate when this treatment system is applied at real field sites, and secondly, these systems in tropical and subtropical areas are especially vulnerable to natural disasters such as hurricanes or typhoons (X. Li et al., 2010; Keizer–Vlek et al., 2014). Although the planted floating–bed system has certain drawbacks, such as it is vulnerable to extreme condition like typhoons, the planted floating reedbed has been

widely used all over the world as an important ecological remediation to control water eutrophication because it is a low–cost, solar–energy based and eco–friendly technology (Hu et al., 2010).

In aquatic macrophyte systems (Figure 2.9), the removal occurs by several mechanisms including: solids settling, plant uptake of contaminants, biotransformation, and physicochemical reactions (Sooknah and Wilkie, 2004). Different biological and physio–chemical processes play a role in nutrient removal using floating treatment wetlands, such as nitrification, denitrification, and phosphorus and heavy metals adsorption (Keizer–Vlek et al., 2014). However, the effect of the vegetation on the overall removal performance is poorly documented (Chang et al., 2013; Van de Moortel et al., 2010). A high number of studies have achieved significant pollutant removal in floating treatment wetlands (e.g., De Stefani et al., 2011; Vymazal, 2007; Keizer–Vlek et al., 2014).

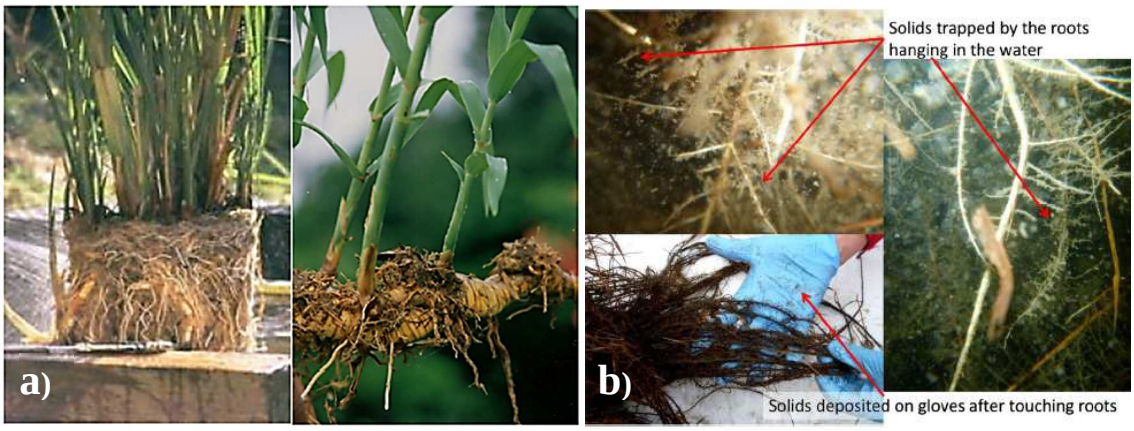


Figure 2.9. Removal of solids by plants in floating treatment wetlands, a) root and rhizome systems of *P. australis* (common reeds) and b) solid partials trapped into biofilm grown on the surface of root and rhizome systems (Borne et al., 2013).

Seasonal effects on floating treatment behaviour can be explained. In summer, the floating treatment wetlands induce an increase in accumulation metals in pond sediment due to lower sediment redox potential, more anoxic water column, neutral pH and greater

source of organic matter. In contrast with the conventional pond sediment, metal release has been detected from the floating treatment wetland pond sediment likely due to aerobic organic matter degradation at the beginning of spring and/or hydroxides reduction when sediments became reduced later in the season (Borne et al., 2014).

A constructed floating mat vegetated with *P. australis* and *Canna indica* showed a decrease in nitrogen by nitrification–denitrification processes, while the phosphorus removal was influenced by filtration and sedimentation. Protozoa predation and oxidation processes affected positively on the removal rates of *Escherichia coli*. Furthermore, the system showed greater removal of nutrients and *Escherichia coli* in the dry period (Saeed et al., 2016).

The common criteria to select wetland macrophytes depend on their availability and abundance in the region of study. Sooknah and Wilkie (2004) selected three floating aquatic macrophytes, namely *Eichhornia crassipes* (water hyacinth), *Hydrocotyle umbellata* (pennywort), and *Pistia stratiotes* (water lettuce), to evaluate the quality improvement of wastewater from anaerobically digested flushed dairy manure, in terms of nutrients, chemical oxygen demand, solids, and salinity. Other free–floating aquatic macrophytes, such as *Ipomoea aquatica Forsskal.* (Swamp morning–glory), *Paspalum repens Bergius.* (water paspalum), *Azolla microphylla Kaulf.* (Mexican mosquito fern), *Salvinia minima Baker* (water spangles), and *Lemna minor L.* (lesser duckweed), have been examined for their effectiveness in treating organic matter and nutrients in wastewater from a dairy farm, a dairy processing plant, a banana paper plant, and a landfill (Nahlik and Mitsch, 2006). Floating treatment wetlands planted with emergent macrophytes, such as *Carex acutiformis, Carex virgata*, *Cyperus ustulatus A.*, *Juncus edgariae*, and *Schoenoplectus tabernaemontani*, have been investigated for their ability to tolerate the wide fluctuations in water depths that are typical of stormwater ponds, as

shown in Table 2.7, (Tanner and Headley, 2011; Van de Moortel et al., 2012). Borne et al. (2013) reported a significantly improved runoff water quality in floating treatment wetlands, vegetated with *Carex virgata*, for remediation of total suspended solids, particulate zinc, and particulate and dissolved copper, compared with parallel stormwater treatment ponds monitored in a field trial study. Furthermore, the floating treatment wetland has shown an increase in adsorption processes for insoluble copper sulphides and direct copper uptake by plants with presence of high humic contents, low dissolved oxygen, and natural water pH. Ladislas et al. (2015) investigated the adsorption mechanism of *Juncus effusus* and *Carex riparia* roots for remediation of cadmium, nickel and zinc from urban water runoff, which demonstrated a high capacity to adsorb dissolved metals and filter particulates (Table 2.7).

An alternative method for selection of vegetation species is to select according to their efficiency of pollutants removal. Chua et al. (2012) reported that *Typha angustifolia* in a floating wetlands system achieved the highest rate of increase in uptake nutrients from the base flow of an urban catchment compared with the other macrophytes such as *Chrysopogon zizanioides* and *Polygonum barbatum*, as shown in Tables 2.7 and 2.8. However, Keizer–Vlek et al. (2014) indicated that nutrient removal values of floating mats planted with *Iris pseudacorus* were significantly higher than the values for total removal from those planted with *Typha angustifolia* and with control wetlands filled with nutrient–contaminated water. Wang and Sample (2014) reported that floating treatment systems with *Pontederia cordata L.* (pickerelweed) showed highest nutrients removal with a considerable phosphorus removal, in particular, compared with system with *Schoenoplectus tabernaemontani* (softstem bulrush). However, the planted systems exhibited a significant removal of nutrients, for both species, compared with control systems without vegetation (Table 2.8).

Table 2.7. Previous studies on treatment of various types of water using floating treatment wetlands (Wang et al., 2014).

Source*	Type of study	Control type	Water type	Coverage[α] (%)	HRT[β] (d)	Air temp., (°C.)	Water temp., (°C.)	Raft area (m^2)	Water depth (m)	Tub dimensions[γ] (m)	Plant density (plant/m^2)
Boonsong and Chansiri 2008	FTW	Open	Primary treated domestic wastewater	56	3–7	**	**	0.72	0.40	L = 0.85, W = 1.50, H = 0.50	83.3
Chang et al. 2012	FTW	Open	Stormwater with added fertilizer	10	15	**	**	0.7 or 1.96	0.56	(1) D = 3.00, H = 0.80 (2) D = 5.00, H = 1.20	**
Hubbard et al. 2004	FTW	None	Swine lagoon wastewater	47	14	20	**	1.00	0.60	D = 1.65, H = 0.91	20–40
Karnchanawong and Sanjitt 1995	Hydroponic	Open	Domestic wastewater	100	1.6–16	**	20–26.9	1.92	0.10	L = 2.40, W = 0.80, H = 1.10	**
Li et al. 2011	FTW	Covered	Urban pond	95	20	20–25	**	0.47	0.15	D = 0.80, H = 1.00	**
Li et al. 2012	FTW	Covered	Refinery wastewater	95	35	0–15	**	0.47	0.15	D = 0.80, H = 1.00	**
Sun et al, 2009	FTW	Open	River water	100	5	**	**	0.96	0.50	L = 1.20, W = 0.80, H = 1.20	**
Van de Moortel et al. 2010	FTW	Open	Domestic wastewater	50	11	3.6–18.3	5.1–20.4	0.77	0.90	L = 1.50, W = 0.80, H = 1.20	**
Van Oostrom, 1995	FTW	Covered	Meat processing wastewater	100	7	**	**	2.88	0.40	L = 2.40, W = 1.20, H = 0.80	**
Wang et al., 2012	FTW	Covered	River water	100 or 25	1–7	**	1–18	2.0 or 0.5	0.60	L = 2.00, W = 1.00,H = 1.00	36
White and Cousins, 2013	FTW	None	Lake water with fertilizer	95	3	**	23–24.8	1.1 or2.9	**	(1) L = 2.10, W = 0.56, H = 0.51; (2) L = 2.50, W = 1.20, H = 0.62	27
Xian et al., 2010	FTW	Covered	Swine farm wastewater	100	7	3–26	10–22	0.2	0.30	L = 0.50, W = 0.40, H = 0.40	80
Yang et al., 2008	Hydroponic	None	River water with dissolved chemicals	100	1–3	15.6	13–19	2	0.60	L = 2.00,W = 1.00, H = 0.75	100
Zhao et al., 2012	FTW	Covered	Urban pond	100	16	12–25	**	0.2	0.60	D = 0.5, H = 0.6	61
Zhou and Wang, 2010	FTW	Covered	River	61	35[δ]	**	14.2–27.6	0.2[θ]	0.12	L = 0.66, W = 0.50, H = 0.38	50
Zhou et al., 2012	FTW	Covered	River	61	48	**	22–30 (summer) 11–16 (winter)	0.2[θ]	0.12	L = 0.66, W = 0.50, H = 0.38	8–10

[α] Coverage is the proportion of water surface area covered by the floating mat.
[β] Hydraulic retention time.
[γ] Length (L), width (W), diameter (D), and height (H).
[θ] Through personal communication with the author(s).
[δ] Purification stage.
* Sources include references cited and Headley and Tanner (2012).
** Not available.

Table 2.8. Mesocosm–scale floating treatment wetlands in different published studies (Wang et al., 2014).

Source[a]	Treatment (plant species, control)	Water		Plants	
		N removal rate (g/m^2 day)	P removal rate (g/m^2 day)	N uptake rate (g/m^2 day)	P uptake rate (g/m^2 day)
Boonsong and Chansiri, 2008	*Vetiveria zizanioides* (L.) Nash	1.1–2.5	0.1–0.88	0.0015–0.0059	0.0057–0.026
Chang et al., 2012	*Canna flaccida* and *Juncus effuses* (mix)	[b]	[b]	0.036	0.0015
Hubbard et al., 2004	*Typha latifolia* L.	1.11	0.16	0.79	0.13
	Panicum hematomon Shult 'Halifax'	0.67	0.10	1.03	0.16
Kamchanawong and Sanjitt 1995	*Ipomoea aquatica*	0.18–0.51	[b]	0.035–0.12	[b]
Li et al., 2011	*Lolium perenne* 'Top One'	0.013	0.004	[b]	[b]
	Lolium perenne 'respect'	0.009	0.004	[b]	[b]
	Lolium perenne 'Top One' and immobilizing denitrifiers	0.014	0.004	[b]	[b]
	Lolium perenne 'respect' and immobilizing denitrifiers	0.011	0.004	[b]	[b]
	Immobilizing denitrifiers	0.008	0.003	[b]	[b]
Li et al., 2012	*Geophila herbacea* O Kuntze	0.057	0.002	0.0016	0.0002
	Lolium perenn cv 'Caddieshack'	0.060	0.002	0.0027	0.0003
	Lolium perenne L.	0.061	0.003	0.0020	0.0002
	Lolium perenne Topone	0.067	0.003	0.0041	0.0005
Sun et al, 2009	*Canna* sp.	0.44	[b]	[b]	[b]
	Canna sp. and immobilizing denitrifiers	0.55	[b]	[b]	[b]
	Canna sp. and aeration	0.64	[b]	[b]	[b]
Van de Moortel et al., 2010	*Carex* sp. (dominant, >95% of the surface area)	0.55	0.02	[b]	[b]
Van Oostrom, 1995	*Glyceria maxima*, normal flow	5.3	[b]	[b]	[b]
	Glyceria maxima, irrigated from top	5.2	[b]	[b]	[b]
	Glyceria maxima, 50% effluent recycled	5.5	[b]	[b]	[b]
Wang et al., 2012	*Phragmites australis* and *Typhus latifolia* and biological ceramic filter substrate, 1–13°C.	22.9	0.5	[b]	[b]
	Phragmites australis and Typhus latifolia and biological ceramic filter substrate, 13–18°C.	66.3	1.8	[b]	[b]
	Phragmites australis and *Typhus latifolia* without substrate, 1–13°C.	10.8	0.3	[b]	[b]
	Phragmites australis and *Typhus latifolia* without substrate, 13–18°C.	52.2	0.9	[b]	[b]
	Phragmites australis and *Typhus latifolia* with fibrous filler substrate, 1–13°C.	11.8	0.3	[b]	[b]
	Phragmites australis and *Typhus latifolia* with fibrous filler substrate, 13–18°C.	61.6	0.8	[b]	[b]
White and Cousins, 2013	*Canna flaccida* and *Juncus effusus* (mixed) 2008	1.05	0.04	[b]	[b]
	Canna flaccida and *Juncus effusus* (mixed) 2009	3.00	0.17	[b]	[b]
	Canna flaccida	[b]	[b]	0.22	0.014
	Juncus effusus	[b]	[b]	0.39	0.024
Xian et al., 2010	*Lolium multiflorum* Lam. 'Dryan'	0.130	0.018	[b]	[b]
	Lolium multiflorum Lam. 'Waseyutaka'	0.092	0.011	[b]	[b]
	Lolium multiflorum Lam. 'Tachimasari'	0.134	0.013	[b]	[b]
	Control	0.103	0.013	[b]	[b]
Yang et al., 2008	*O. javanica*	2.740	0.010	[b]	[b]
		0.220	0.030	[b]	[b]
		0.07	0.037	[b]	[b]
Zhao et al., 2012	*Zizania caduciflora*	0.05	[b]	[b]	[b]
	Triarrhena lutarioriparia	0.15	[b]	[b]	[b]
	Thalia dealbata	0.13	[b]	[b]	[b]
	Vetiveria zizanioides	0.15	[b]	[b]	[b]
	Miscanthus sinensis Anderss sp.	0.16	[b]	[b]	[b]
	Acorus calamus	0.18	[b]	[b]	[b]
	Control	0.14	[b]	[b]	[b]
Zhou and Wang, 2010	*Oenanthe javanica* 'Blume'	0.0465	0.0021	2.47	0.38
Zhou et al., 2012	*Rumex acetosa* 'Linn.', summer	0.060	[b]	2.8	[b]
	Rumex acetosa 'Linn'., winter	0.057	[b]	2.4	[b]

[a] Sources include references cited and Headley and Tanner (2012).
[b] Not available.

Furthermore, Wang et al. (2014) have recommended harvesting pickerelweed foliage to maximise nutrient removal at the end of summer, because it translocates most of its nutrients to below–ground storage organs in the autumn, resulting in less nutrient mass in the above–ground tissue compared to the case in the summer. Since, pickerelweed has shown high growth biomass with great nutrient removal, followed by softstem bulrush in almost reported findings. Softstem bulrush and yellow iris can be sustained over winter. In general, harvest of above–ground vegetation at the end of the growth season was recommended to maximise nutrient removals (Wang et al., 2015).

Considering plant species in subtropical stormwater wet detention ponds, nutrient uptake and assimilation by *Juncus effusus* (soft rush) was much higher than that by *Pontederia cordata* (pickerelweed) through both leaves and roots (Chang et al., 2013). While, White and Cousins (2013) concluded that both floating treatment wetlands planted with *Canna flaccida* and *Juncus effuses* have proven most effective in low nutrient environments, where it is necessary to polish water quality to extremely low phosphorus concentrations. Furthermore, there are no significant differences between the two species in terms of removal capacity at two rates (low and high) of nutrient loading to simulate normal– and nutrient–rich runoff of stormwater loading conditions. Despite the combination with biofilm bacterial community in floating treatment wetlands, *Alternanthera philoxeroides* and *Cyperus alternifolius* have recorded greater removals of total phosphorus, ammoniacal nitrogen, and biochemical and chemical oxygen demand. *Scirpus validus* has exhibited significant reduction of total nitrogen and *Canna generalis* removed nitrate–nitrogen when treating wastewater (C. Zhang et al., 2014).

The size and relative surface cover of the floating treatment wetland, the relative root depth, and the capability of the plants to tolerate periodic anaerobic conditions have been indicated as crucial factors for promotion of a good removal across a spectrum of

pollutants (Borne et al., 2015). The greater coverage area of floating macrophytes for the pond was proven to improve pollutants removal by Winston et al. (2013). In that study, two constructed floating wetlands were covered with 9% and 18%, respectively, with a mixture of *Carex stricta* (tussock sedge), *Juncus effusus* (soft rush), *Spartina pectinata* (prairie cordgrass), *Acorus gramineus* (Japanese sweet flag), *Pontederia cordata* (pickerelweed), *Peltandra virginica* (arrow arum), *Andropogon gerardii* (big bluestem), and *Hibiscus moscheutos* (marsh hibiscus), (Winston et al., 2013). But, Chang et al. (2012) argued that the application of floating vegetated islands as a stormwater technology can remove nutrients through plant uptake and biological activity and the most cost–effective size in the outdoor mesocosms to be 5% surface area coverage of the pond surface. In addition to coverage surface area, orientation and arrangement of floating reedbeds have an essential effect on removal efficiency.

Khan et al. (2013) improved the performance of a stormwater retention pond when floating treatment wetlands (vegetated with *Carex virgata*, *Cyperus ustulatus A.*, *Juncus edgariae*, and *Schoenoplectus tabernaemontani*) were positioned centrally across the width of the pond.

Furthermore, Tanner and Headley (2011) concluded that biofilm associated with submerged root has a key role in the removal of copper, dissolved reactive phosphorus, and fine suspended particles compared to the control of non–planted floating mats. Three pollutant–degrading bacteria (*Acinetobacter sp.*, *Bacillus cereus*, and *Bacillus licheniformis*) inoculated with *Brachiaria mutica* vegetated floating treatment wetlands maximised the removal rates of nutrients and biochemical and chemical oxygen demands from wastewater. Both organic and inorganic contaminants were potentially removed by detoxification of sewage effluent produced by *B. mutica* bacteria (Ijaz et al., 2015).

Further studies have considered different strategies to enhance the performance of floating treatment wetlands. One example, used ecological sludge produced by utilisation of dredged lake sludge as a solidification/stabilisation treatment combining with a macrophytes–based treatment of *Acorus calamus* for promising removal efficiency of nitrogen, phosphorus and *chlorophyll–a* (Hu et al., 2010). Furthermore, addition of immobilised denitrifying bacteria and aeration into the floating treatment wetlands enhanced the nitrogen removal efficiency, and the removal of chemical oxygen demand as well, from polluted river water in five days of treatment (Sun et al., 2009).

In addition, an integrated ecological floating–bed artificial ecosystem was developed by introducing both a biofilm carrier, functioning as a medium for biofilm growth, and *Corbicula fluminea* (Asian clam), a kind of freshwater bivalve, into the conventional planted floating–bed (*Ipomoea aquatica*) to improve the performance of the treatment system. The system proved its ability for purification of the water by increasing the removal percentages of nutrients, organic matters, and microcystin–LR with a 7– day water exchange period (X. Li et al., 2010). Enhancement of the performance of an integrated floating island system was achieved by using a mosaic floating island with adsorptive biofilms planted with *Thalia dealbata* (alligator flag) to mitigate nutrients, *chlorophyll, Escherichia coli* and heavy metals from eutrophic water river (Zhao et al., 2012).

An aerator device operated by solar power was applied to an artificial floating island cultivated with *Typha orientalis Presl, Eleocharis dulcis* and *Juncus effuses* to treat runoff water and domestic wastewater. The results reflected an increase in dissolved oxygen and oxygen reduction potential, high removal and breakdown of nutrients, and inhibition of algae growth (Lu et al., 2015). Two commercial batch–fed mesocosms of Beemats and BioHaven floating islands cultivated with *Juncus effusus* (soft rush) were

investigated by removing nutrients from runoff of a combined irrigation holding and stormwater retention pond. Due to the differences between mat material or substrate materials and/or additives of the two treatments, nutrient removal in the BioHaven was indicated to be lower than in the Beemat treatment over the entire experimental period, with a 7–day retention time. However, the two technologies showed similar nutrient removal during the plant growth season. So, the Beemat mat yielded a significant positive net removal of nutrients, while the BioHaven had a significantly greater biomass. Furthermore, both floating treatment wetlands showed greater biomass accumulation in shoots rather than in roots (Lynch et al., 2015).

2.4.3. Pollutant Removals in Wetlands

Engineering the constructed wetland treatment systems to optimise the removal mechanism for pollutants is required for a particular wastewater (Reddy and D'angelo, 1997). Macrophytes, microorganisms, and substrate soil are the essential compounds of reedbed treatment systems (Figure 2.10). The role of macrophytes is essential for providing a surface and environment for microbial growth and filtration as well as the substrate surfaces (Brix, 1997).

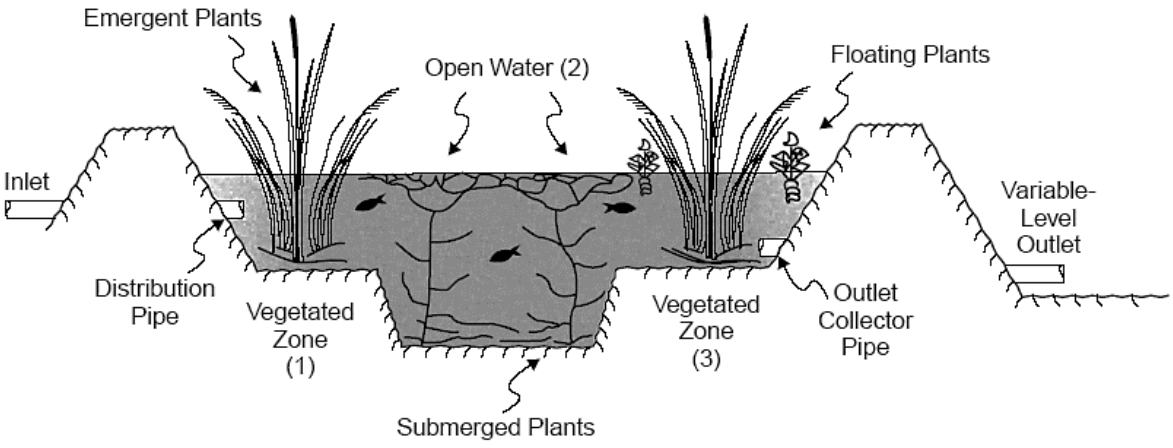


Figure 2.10. Types of macrophytes in constructed wetlands (USEPA, 2000).

In addition, their roots play a vital role in transferring oxygen to the rhizosphere for microbial population (Brisson and Chazarenc, 2009). So, from a biological point of view, the reedbed treatment systems are far more diverse than conventional biological treatment systems. However, contaminates are removed in wetlands in a complex variety of processes which could be physical, chemical, and biological as well (Brix, 1993).

In the presence of dissolved oxygen, organic constituents in wastewater are degraded aerobically by bacteria associated with wetland macrophytes and the matrix surface of wetlands (Vymazal, 2005). The required oxygen is diffused directly from the atmosphere to the rhizosphere by plant roots. While, anaerobic degradation can be indicated with the absence of dissolved oxygen (Brix, 1987). The anaerobic stage is considered an important phase and predominates for specific wastes, especially those originating from organically overloaded systems. Uptake of organic matter by macrophytes in wetland system is a much lower portion compared with biological processes (Vymazal, 2007).

Organisms must have a source of carbon for synthesis of new cellular material, energy, and, whether organic and/or inorganic, nutrients are required. Insufficient nutrient contents in some effluents, such as industrial wastewater, leads to inefficient biological treatment (Elser et al., 1996).

In general, light and chemical oxidation–reduction reactions are the main sources of energy which can be driven by organisms for cell synthesis, which are known as phototrophic and chemotrophic, respectively. Furthermore, carbon sources for microorganisms could be either from organic chemicals, for heterotrophic organisms, or from carbon dioxide, for autotrophic organisms (Zhang et al., 2003). Therefore, microorganism groups that use organic carbon for the formation of cell tissues with light

or organic oxidation–reduction reactions as a source of energy are known as *photo–heterotrophic* (sulphur bacteria) or *chemo–heterotrophic* (protozoa and most bacteria), respectively. While, the groups that derive cell carbon from carbon dioxide by obtaining energy from light or oxidation of reduced inorganic compounds, such as ammonia and nitrite, are called *photo–autotrophic* (photosynthesis bacteria) or *chemo–autotrophic* (nitrifying bacteria), respectively. For example, the *heterotrophic* organisms are the primary concern in treatment with a major objective to remove organic matter because of their need for organic material as a source of carbon in metabolism and respiration (Wagner et al., 1996). With presence of oxygen (Figure 2.11), soluble organic chemicals are degraded aerobically by *chemoheterotrophic* microorganisms which oxidise organics to ammonia, then the ammoniacal nitrogen will be oxidised to nitrite and nitrate by *chemoautotrophic* process of nitrification (Reddy et al., 1989).

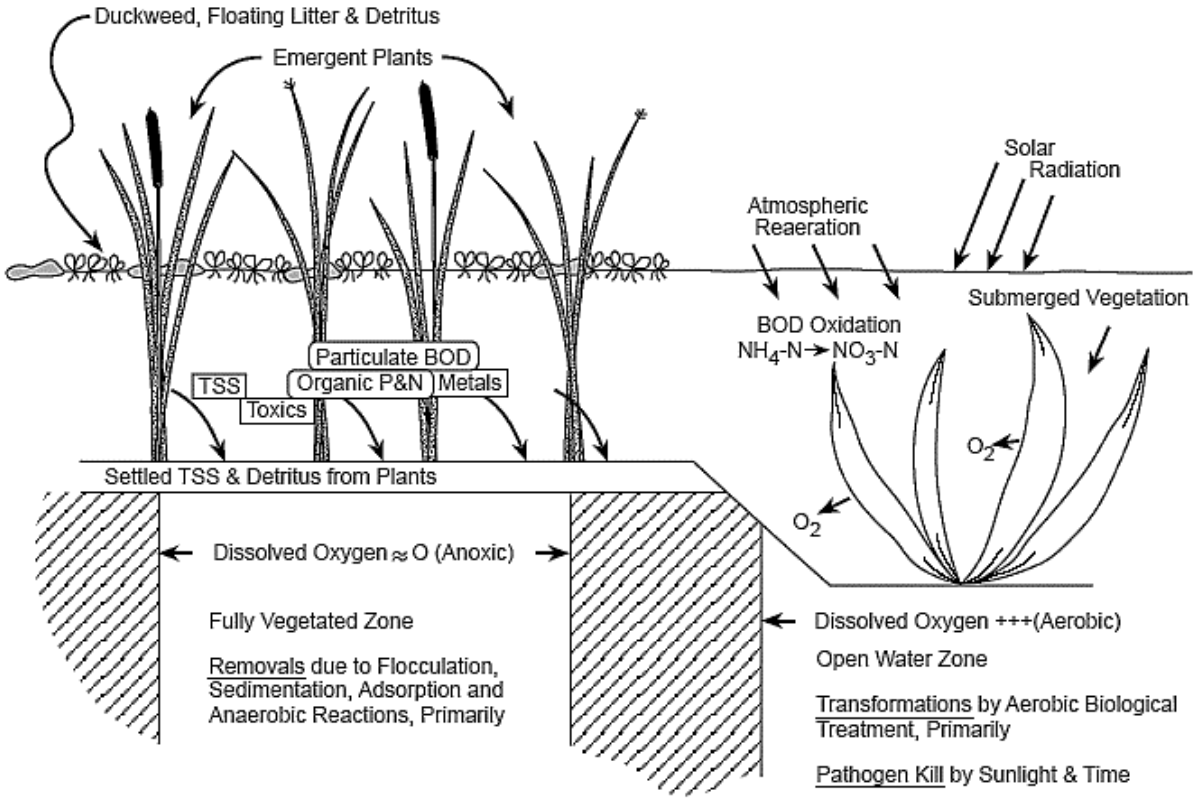


Figure 2.11. Oxygen transformations in constructed wetlands (USEPA, 2000).

In this case, the heterotrophs are mainly responsible for consuming organics, due to their faster rate of metabolism, in the biochemical oxygen demand of the system. The performance of aerobic biological oxidation in wetlands depends of the availability of oxygen and the amount of active organic matter for microorganisms. In the absence of dissolved oxygen, the overall description of anaerobic degradation is a biochemical process to digest organic matter by anaerobic *heterotrophic* bacteria into carbon dioxide and methane (Figure 2.12). This process occurs in constructed reedbed wetlands through four stages (Amon and Benner, 1996).

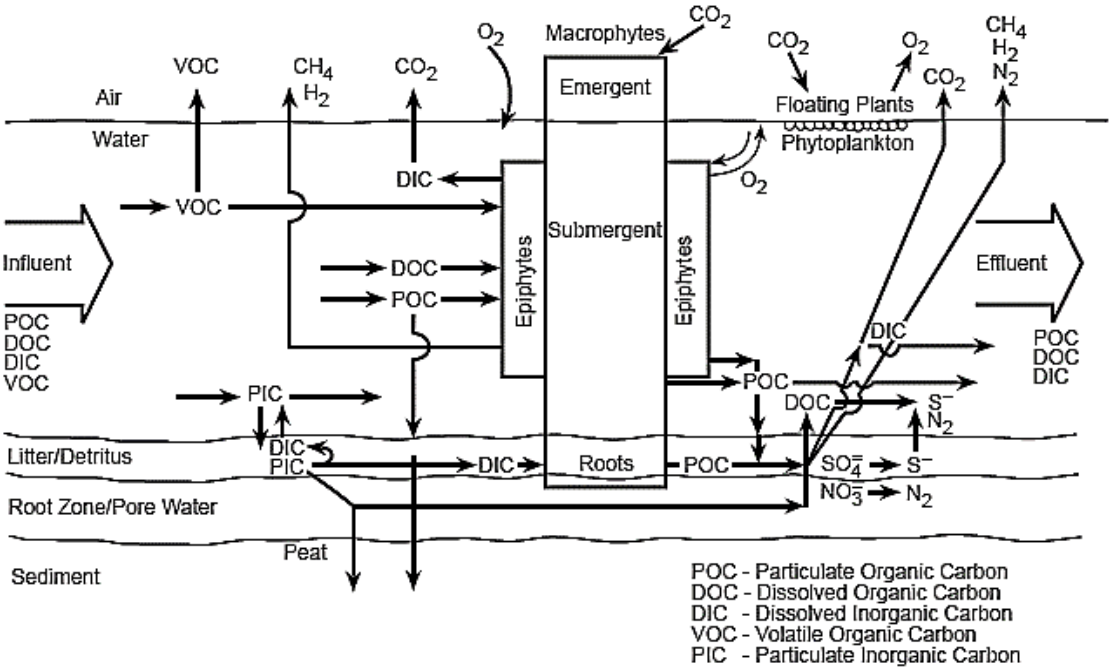


Figure 2.12. Carbon transformations in constructed wetlands (USEPA, 2000).

Firstly, the anaerobic bacteria access the potential energy of the biomass which is made up of large organic polymers. The acid–forming bacteria must first be broken down into their smaller constituent parts. The breaking process and dissolving the smaller molecules into solution is called hydrolysis. Through hydrolysis, the complex organic molecules are broken down into simple sugars, amino acids, and fatty acids. Acetate and hydrogen are also produced in this stage, but they are available for methane–forming

bacteria. However, long chain molecules which are also present in the first stage, such as volatile fatty acids, must be catabolised into their simple compounds to be directly used by methanogens (Thauer et al., 1977).

Secondly, acidogenesis is further breakdown of the remaining components as a result of the biological process of acidogenic (fermentative) bacteria. In this stage, volatile fatty acids are created along with ammonia, carbon dioxide, and hydrogen sulphide, as well as other by–products. The third stage of anaerobic degradation is acetogenesis. Simple molecules created with further digestion by acetogens to produce largely acetic acid, as well as carbon dioxide and hydrogen (Ghosh et al., 1975).

Methanogenesis is the final stage of anaerobic degradation. The biological process of methanogens uses the intermediate products of the preceding stages and converts them into methane, carbon dioxide, and water. These components make up the majority of the biogas emitted from the system. Methanogenesis is sensitive to both high and low pH and occurs between pH 6.5 and pH 8. Stopping of methane production normally occurs in anaerobic degradation due to over–production of acid, which leads to rapidly decreasing pH with release of the foul odour of hydrogen sulphide from the reedbed treatment system (Thauer, 1998).

Nitrification and denitrification are the main processes of the removal mechanism of nitrogen in wetland treatment systems (Figure 2.13). Volatilisation and adsorption are also other mechanism of removal but they are generally less importance. The nitrification is a chemoautotrophic process where carbon dioxide is used as the source of carbon and oxidation of ammonia is the source of energy. In aerobic zones, nitrifying bacteria oxidises ammonia or ammonium to nitrite by Nitrosomonas followed by oxidation of nitrite to nitrate by Nitrobacter (Brix, 1993; Vymazal, 2007).

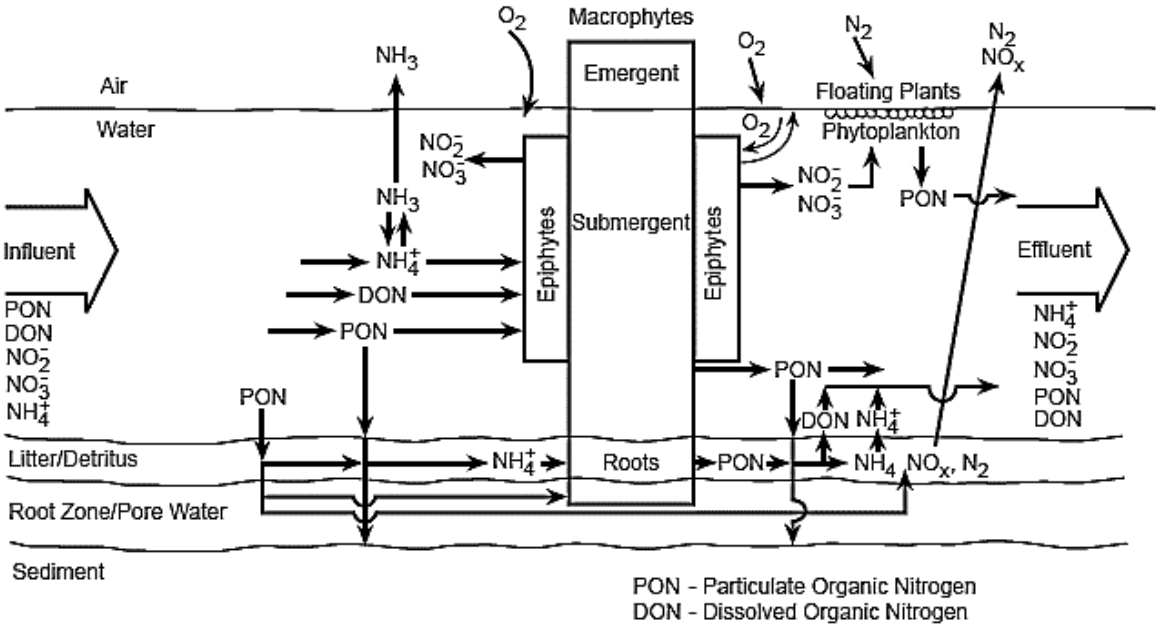


Figure 2.13. Nitrogen transformations in constructed wetlands (USEPA, 2000).

Sensitivity of nitrifying bacteria is mainly affected by a wide range of inhibitors such as high concentrations of ammonium and a narrow range of pH between 6.5 and 8.6 with a minimum temperature for growth of Nitrosomonas of 5 °C and Nitrobacter of 4 °C. Dissolved oxygen is also required at a concentration above 1 mg/l, where approximately 4.3 mg of oxygen is needed to oxidise ammoniacal nitrogen to nitrate nitrogen (Wett and Rauch, 2003).

Then, the nitrate will be converted to dinitrogen gas by denitrifying bacteria, mostly chemoheterotrophic, in two stages within anoxic zones. The first stage is the conversion of nitrate to nitrite, followed by the second stage to convert nitrite to nitric oxide, nitrous oxide, and nitrogen gas. The process to mineralise organic nitrogen, in both aerobic and anoxic zones of reedbed treatment systems, to ammoniacal nitrogen is called ammonification or mineralisation (Vymazal, 2007).

In the denitrification process, the presence of oxygen suppresses the enzyme system. Alkalinity is produced in denitrification which can result in increased pH, which

has required limits between 7 and 8, however, alkalinity is consumed in the nitrification process. Organic carbon is a critical parameter that should be available in wastewater or the bed matrix to convert process of denitrifying bacteria at a minimum temperature of 5 °C.

Organic and inorganic forms of phosphors are naturally present in wetlands. In wastewater, the biologically available form of phosphorus is typically an ortho–phosphate–phosphorus referred to as soluble inorganic phosphorus (Boström et al., 1988). The other forms, such as insoluble organic and inorganic and dissolved organic phosphorus, are non–reactive until converted to orthophosphate form by biological oxidation. There are two main mechanisms for removal of orthophosphate from wastewater within wetlands (Brix et al., 2001), either as a sequence of adsorption, complexation, and precipitation reactions with ferric iron, calcium, and aluminium found in wetland soils, and/or as a sequestration in living biomass into wetlands (Figure 2.14).

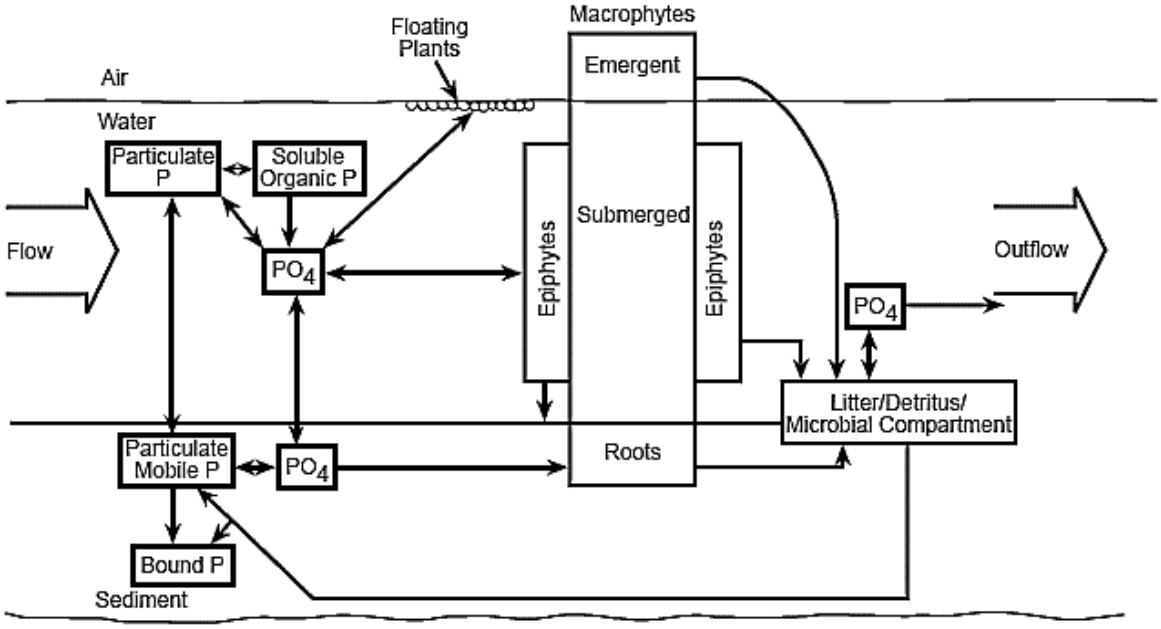


Figure 2.14. Phosphorus transformations in constructed wetlands (USEPA, 2000).

There is a symbiotic relationship between macrophytes and bacteria within wetland systems whereby the provided surface area and diffused oxygen by the root system of plants are necessary for predominating bacteria. The majority of bacteria indicated in floating treatment wetland environments are heterotrophic bacteria with much fewer autotrophic species (Kadlec and Wallace, 2009). Autotrophic bacterial species also play an important role in pollutant removal through nitrogen removal. *Nitrosomonas* and *Nitrobacter* which are *chemoautotrophic* bacteria, oxidise ammonia to nitrite and nitrite to nitrate, respectively (Cooper et al., 1996; Kadlec and Wallace, 2009). Through the complex processes of degradation, organic matter broken down due to bacterial activities could be available for uptake or volatilisation from the water column by plant roots leading to increased removal efficiency (C. Zhang et al., 2014).

However, some aquatic macrophytes release nutrients back to the water body at the end of growth seasons, but the removal reaches its peak at the initial stage of growth, such as in *P. australis* and *Typha latifolia* (Kröger et al., 2007). So, harvesting foliage parts is a possible solution to prevent release of pollutants (Borne et al., 2015). Another promising contribution of the macrophytes root system is trapping suspended particles, which can be nutrients and metals, when wastewater passes through. This mechanism can also release or re–suspend particles either under turbulent water flow and or sloughing off the heavy biofilm created by attachment of particles and dead bacteria (Kadlec and Wallace, 2009; Tanner and Headley, 2011). In addition, root systems can reduce water velocity, resulting in more chance to settle the sloughed off bio layers and suspended particles, which are organic or inorganic particles, and incorporate them into the wetland bottom (Dunne et al., 2012).

Coverage surface area provided by floating mats can control the dissolved oxygen, water temperature and evaporation (Van de Moortel et al., 2010). Reducing

sunlight penetration to a water body might prevent algae growth and consequently decrease algae die off (Headley and Tanner, 2012).

Oxygen depletion due to excessive growth of algae and sediment bacteria, which consumes oxygen to degrade dead algae (Lee and Lee, 1995), could also be mitigated within water bodies of floating treatment wetlands. However, a disadvantage appears which a reduction in the interface area between air and water which affects the quantity of diffused oxygen going into the water column, but this oxygen could be substituted by the roots of macrophytes which are cultivated in the floating mat of wetland (Tanner and Headley, 2011).

2.4.4. Wetlands for Greywater Treatment

The constructed wetland has been considered as the most environmentally friendly and cost–effective technology for greywater treatment (F. Li et al., 2009).

A pilot–scale recycled vertical–flow constructed wetland was studied for populations of various species of organisms in three different formulations of synthetic greywater to represent the variable composition of domestic greywater. These results underscore the need for extensive microbiological monitoring of greywater systems as well as the need for the development of greywater specific standards for the possible reuse of greywater for irrigation (Gross et al., 2006).

Real domestic greywater with low–organic strength was treated by different designs of constructed wetlands, namely horizontal sub–surface flow wetland, vertical flow wetland, and green roof water recycling system. The first two wetlands were vegetated with *P. australis* while the latter was planted with different marginal plants. The conducted results were compared in terms of biochemical oxygen demand, total suspended solids, turbidity, and pathogens (Avery et al., 2007).

An experimental system of a recycled vertical–flow bioreactor was examined for the removal of chemical and microbiological contaminants from synthetic greywater enriched with wastes from a dining hall. The system reduced the chemical pollutants to the acceptable limits for either recreation or irrigation associated with a significant increase in the number of both heterotrophic bacteria and surfactant–degrading bacteria within the system. The system also resulted in final pathogen contaminants that met the USEPA water quality criteria for recreational water (Gross et al., 2007a).

The recycled vertical–flow constructed wetland was concluded as an economically sound, low–tech and easily maintainable treatment system that would allow safe and sustainable use of greywater for landscape irrigation in small communities and households. Removal efficiency, in terms of suspended solids and biological, and biochemical and chemical oxygen demand, hydraulic parameters and feasibility were studied, as well as the environmental effects of the treated greywater, as reflected by soil and plant parameters over time (Gross et al., 2007b).

The vertical–flow wetland was the most reliable performer under low– and high–strength influent conditions of synthetic greywater, indicating aerobic unsaturated wetland to be the most suitable form of the technology for pathogen removal. This conclusion was drawn following comparison of three different constructed wetlands, a membrane bioreactor, and a membrane chemical reactor (Winward et al., 2008)

A pilot–scale ecological sanitation system was implemented to evaluate the efficiency of the biofilters and a horizontal sub–surface flow constructed wetland, vegetated with two species of terrestrial ornamental plants, to treat domestic greywater. The combined system resulted in an increase in dissolved oxygen with high removal of nutrients and physiochemical parameters. The biofilters contributed most of the total

removal in terms of biochemical and chemical oxygen demand, however, the wetland and biofilters were equally efficient in total suspended solids removal. Both species of plants grown in the wetland contributed to removal of pathogens and phosphorus from greywater (Ling et al., 2009).

A controlled study utilising fresh water, raw artificial greywater or treated artificial greywater demonstrated that treated greywater with a recirculating vertical–flow constructed wetland can be effectively used for irrigation without detrimental effects on soil or plant growth in terms of physicochemical and biological parameters compared with raw GW that can impact the movement of water in soil and the transport of contaminants in the vadose zone (Travis et al., 2010).

A functional hybrid phytoremediation pilot platform was studied to evaluate its feasibility and treatment efficiency under variable stress conditions. The pilot plant was tested with and without vegetation at the design specifications and different organic and hydraulic loads of greywater were applied. The hybrid system consisted of two vertical sub–surface flow wetlands working in parallel, followed by a horizontal sub–surface constructed wetland with the possibility of a recycle loop. The system showed its ability to reduce the chemical oxygen demand over the treatment stages and over the loading rates (Comino et al., 2013).

To assess the environment and human health risks associated with on–site treatment and reuse of greywater for irrigation purposes, vertical–flow and recycling vertical–flow constructed wetlands were occupied to treated domestic greywater. The water samples were analysed for physicochemical and quantitative microbial risk assessment which was affected by type of wetland design, type of greywater source, and the associated microbiological loads (Maimon et al., 2014).

A greenhouse laboratory–scale experiment was established to demonstrate the performance of free water surface–flow constructed wetland vegetated with *P. australis* (common reed) operated to treat domestic kitchen wastewater. The investigation recorded a significant reduction in nutrients and other pollutants and no significant change in total dissolved solids. However, an increase was indicated in pH, magnesium, iron, and calcium. Therefore, the effluent properties were concluded to be suitable for non–potable uses such as crop irrigation or direct discharge to water bodies (Oladejo et al., 2015).

A prototype wetland system was evaluated to remove various contaminations from synthetic greywater by the root zone treatment process using Heliconia angusta. A significant removal from outflow discharge was achieved in terms of nutrients, microorganisms, and heavy metals as compared with standard specifications (Saumya et al., 2015).

In general, greywater is generated as a result of the living habits of the people involved, the products used and the nature of the installation and, therefore, its characteristics are highly variable (Eriksson et al., 2002). The major difficulty present in the treatment of greywater is the large variation in its composition (Al–Hamaiedeh and Bino, 2010). However, greywater tends to be more lightly polluted than blackwater, particularly in terms of suspended solids and pathogens (Jefferson et al. 1999) and it therefore should require less extensive treatment prior to reuse.

2.5. Phosphorus Removal in Wastewater Treatment Plants

Different types of wastewater are essentially treated either for recycling or for discharging to the environment (Asano and Levine, 1996). The sensitive environmental concern is aquatic eutrophication and toxic algal blooms which are linked to diffusion of nutrients such as nitrogen and phosphorus, in particular, from municipal, industrial and

agricultural wastewaters (Anderson et al., 2002). Furthermore, agricultural wastewater is considered as a major source of phosphorus in wide applications such as leaching phosphorus–based fertilisers from irrigation water, animal farm wastes disposal and large–scale animal feeding operations (Carpenter et al., 1998; Correll, 1998; Daniel et al., 1998; Ulén et al., 2007). Therefore, phosphorus concentration is limited by many legislations in household products and in the treatment requirements of municipal and industrial wastewaters in order to minimise the daily load of the released phosphorus from these sources (Parry, 1998; Neeteson, 2000; Meinzinger and Oldenburg, 2009).

Particulate, soluble and total phosphorus forms are normally measured in investigation wastewater quality before and after treatment. In traditional primary and secondary treatment plants, particulate phosphorus was mainly removed (Metcalf et al., 1991). Because orthophosphate is primary fraction of the soluble phosphorus form responsible for eutrophication, tertiary treatment of secondary effluents designates for removing soluble phosphorus to meet the international environmental requirements (Dueñas et al., 2003). Long–term sustainability, efficiency and economy are significant required aims for treatment technologies to decrease the flow of pollutants into the environment and to mitigate the phytoplankton growth, which consumes orthophosphate, causing eutrophication (Abell, et al., 2010).

In biological treatment processes of wastewater, phosphorus is an essential macronutrient to assist microorganism growth and phosphorus concentrations could be governed when elevated in wastewater. Biological treatment can also involve phosphorus removal through taken up, with increasing growth of bacteria inhabitants in an anaerobic/anoxic (Morse et al., 1998). The required steady supply of carbon is often an issue in operation of biological treatment systems, without it biological treatment will be

temperamental and may be insufficient to consistently remove phosphorus at a low carbon concentrations (Morse et al., 1998; Seviour et al., 2003).

However, it is common practice to add aluminium or iron salts in a tertiary treatment stage for phosphorus removal. A heavy floc blanket forms and settles down through the water when aluminium or iron salts, such as alum, are added to the wastewater in typical treatment systems (Licsko, I., 1997). Usually, a combination of mechanisms leads to the removal of phosphorus from wastewater, namely adsorption by the aluminium or iron oxide floc, and direct precipitation of aluminium or iron phosphate. Adding chemicals two or three times could achieve complete removal of phosphorus (Yeoman et al., 1988).

In addition, substances containing calcium, such as lime, are utilised to achieve good phosphorus removals served with a high level of pH, as this is more economic especially for large volumes of discharge (Yeoman et al., 1988).

There is a wide range of suggested technologies for wastewater treatment, mainly phosphorus removal, based on physical separation, chemical precipitation, biological uptake, adsorption processes, and/or or a combination of two or more approaches, such as in various types of constructed wetlands. However, large capital investment is required and high costs may be incurred for operation and maintenance of infrastructure and reagent chemicals for such technologies (Morse et al., 1998; Kivaisi, 2001).

To reduce consumption of commercial reagent chemicals for phosphorus removal in treatment of wastewater, it was suggested to use raw minerals and waste by–product industrial materials as sludge–based technologies for P sequestration. Therefore, it is desirable to develop phosphorus adsorption using iron oxide/hydroxide and aluminium hydroxide materials which has been recently studied and found to be successful

(Wieserman et al., 1991; Heal et al., 2003; Wei et al., 2008; Bhatnagar and Sillanpää, 2010).

2.6. Phosphorus Removal by Fly Ash

Fly ash as a waste material, originating from combustion of coal in power stations, has been considered as a phosphorus adsorbent due to its high contents of silicon, aluminium, iron, calcium and magnesium in oxide forms (Oguz, 2005; Chen et al., 2007; Lu et al., 2009). In previous research, the majority of phosphorus was rapidly removed within the first five minutes due to the high pH value and calcium content which both decreased the phosphorus content by precipitation followed by a slow adsorption mechanism (Ugurlu and Salman, 1998; Lu et al., 2009). However, it has been reported that calcium and iron contents of fly ash correlate strongly which could indicate that both precipitation and adsorption are important mechanisms for phosphorus removal (Chen et al., 2006). The pH has been indicated as a governing factor for the removal of phosphorus from aqueous solution by fly ash (Chen et al., 2007), where low and high values of pH could contribute to release and disband calcium, respectively, leading in both cases to retard the removal of phosphorus (Can and Yidliz, 2006). Therefore, a neutral pH was decided to be the most advantageous, however, pH values of 9 and above are generally required for efficient calcium phosphate precipitation (Johansson and Gustafsson, 2000).

2.7. Phosphorus Removal by Slag

Slag is presented in steel–making industries as a waste by–product material with variable chemical composition depending on the raw materials and process used for the steel production. It has been utilised to remove phosphorus from solutions because it contains oxides of calcium, magnesium, aluminium, silicon and iron (Xiong et al., 2008; Xue et al., 2009). The phosphorus adsorption capacity of slag has been reported to vary

depending on its chemical composition, crystalline structure and test conditions amongst other factors. It has been claimed that the adsorption mechanism is dominant, followed by precipitation (Kostura et al., 2005). Through increasing the pH level, a decrease in phosphorus removal by slag was observed due to changes in the slag surface charge from positive to negative causing repulsion with the negatively charged phosphate ions (Xue et al., 2009). Furthermore, removal of phosphorus could occur through precipitation of calcium, magnesium and aluminium in the form of phosphates (Xue et al., 2009), especially when dosing phosphate and monitoring the decrease in the dissolved calcium and pH in the aqueous solution, simultaneously (Johansson and Gustafsson, 2000).

In contrast, a positive correlation between high level of pH and phosphorus removal was stated (Kostra et al., 2005; Kim et al., 2006), and pH values of above 9 and large amounts of soluble calcium which is released from the slag into the solution are required for effective phosphate removal (Johansson and Gustafsson, 2000). Therefore, calcium to phosphorus molar ratios have been reported to achieve maximum phosphorus removal by the precipitation mechanism at high pH values, regardless of the condition of low pH values for favouring adsorption (Johansson and Gustafsson, 2000; Kim et al., 2006; Drizo et al., 2006).

2.8. Phosphorus Removal by Concrete Waste

Fly ash and/or slag have been blended with Portland cement to investigate their potential abilities for phosphorus removal (Agyei et al., 2002). It has been indicated that precipitated calcium phosphate reveals the correlation between the calcium oxide content of the cement–waste blend and the calcium ion released into solution. So, removal of phosphorus by only cement is greater than by the blended substances (Oğuz et al., 2003; Berg et al., 2005; Park et al., 2008). However, Agyei et al. (2002) reported that

phosphorus removal has an inverse correlation with pH and a slight increase in phosphorus removal was observed above a pH value of 9.

2.9. Phosphorus Removal by Animal Waste

Animal wastes which contain relatively high calcium, such as oyster shells and cow bones, have been studied in removal of phosphorus experiments (Lee et al., 2009; Yu et al., 2010; Jang and Kang, 2002). Phosphorus removal was shown to be highly correlated with increasing calcium ion concentration and pH.

Several other studies utilised waste materials for phosphorus removal, such as fly ash from coal combustion (O'Reilly and Sims, 1995), steel–making slags (Drizo et al., 2006), sedimentations of water treatment plants (Makris et al., 2004), or granulated residuals from membrane bioreactor effluents (Genz et al., 2004) for their calcium, aluminium, silicon and iron oxides and/or hydroxides (Gustafsson et al., 2008). However, almost all studies concluded that ferric hydroxides show a higher capacity than alumina for phosphorus removal (Genz et al., 2004).

Furthermore, phosphorus sorption media containing iron oxide nanoparticles embedded in an anion exchange resin was introduced by Blaney et al. (2007). It was expected that waste materials may not be readily available in many geographical areas, and transport costs could prevent their utilisation, although they showed high efficiencies for phosphorus removal. Therefore, ochre is suggested as an alternative source of iron and aluminium oxides which is generated by the neutralisation of acid minewater drainage (AMD) from coal and metal mining regions.

2.10. Phosphorus Removal by Minewater Drainage Sludge (Ochre)

There has been a wide interest in recycling and reuse of industrial waste sludge such as ochre for remediation of various types of wastewater, particularly to adsorb phosphorus (Heal et al., 2003). Ochre is a by–product of mineral sludge with a majority of hydrated ferric oxide ($Fe(OH)_3$) obtained from the drainage of disused coal mine works (Hancock, 2005). Ochre consists predominantly of the same hydroxides, such as aluminium (e.g., alum) or iron salts (e.g., ferric chloride), applied in traditional wastewater treatment processes for the removal of phosphorus (Sibrell et al., 2009). Furthermore, the cost of the chemical reagents used for the treatment of wastewater are relatively high. Both disposal and stockpiling of ochre sludge is costly. The annually generated ochre in the United Kingdom (UK) is around 32,000 tonnes. In comparison, the cumulative sludge in England is only about 19,000 tonnes/annum (Hancock, 2005; Dobbie et al., 2009). Thus, offsetting costs by exploring the feasibility of reusing ochre to improve wastewater quality is highly attractive.

Among various wastewater treatment technologies, constructed wetlands have been considered as a suitable best management practice for treating wastewater economically (low construction, operational and maintenance costs), energy–efficiently (natural physical, chemical and biological–based processes) and in an environmentally–friendly manner compared to other more traditional civil and mechanical engineering approaches. However, the removal of phosphorus in constructed wetlands is not efficient, because of the deficiency in terms of adsorbing and retaining this element (Scholz and Lee, 2005). Wetlands are also often low in Al and Fe. Although phosphorus is vital for the growth of aquatic plants and microorganisms, too much leads to eutrophication of natural watercourses. Thus, the utilisation of ochre, as an alternative source of iron and

aluminium oxides, is highly recommended to increase the phosphorus removal in wetland systems (Heal et al., 2003; Sibrell et al., 2009; Dobbie et al., 2009).

Because of its fine–grained texture and low hydraulic conductivity, ochre is unsuitable for use as a substrate for filter units or as wetland media. Difficulties, such as rapid clogging of treatment systems when using raw ochre sludge, have led to the creation of ochre in the form of granular pellets by mixing it with Portland cement (Heal et al., 2005; Dobbie et al., 2009; Sibrell et al., 2009; Carr 2012; Littler, 2012). Cement–ochre pellets are robust solids, easy–to–handle, linked to a high hydraulic conductivity and characterised by a high absorbent reactivity compared to raw ochre sludge. According to Littler et al. (2013), pelletised ochre was found to remove phosphorus in tests for up to seven days. The pellets are characterised by high removal capacities and operational lifetime. In comparison, the unbound mine sludge seemed to reach its equilibrium within a phosphate solution after about 60 minutes of contact time (Littler et al., 2013). Pollutant removal from wastewater in the presence of cement–bound ochre may occur by physical precipitation under alkaline conditions, although this mechanism is considered less effective than removal by chemical sorption (Heal et al., 2005).

Phosphorus is present in domestic greywater, which is generated from household washing activities at varying concentration levels due to the use of a wide range of detergents (Revitt et al., 2011). So, the use of ochre in wetlands for greywater treatment is an option (Gross et al., 2007b).

2.10.1. Formation of Ochre Sludge

In the late twentieth century, the majority of the UK's metal and coal mines were abandoned. Subsequently, rising of the water table from those deep mines, which was under control during the operation, causes oxidation of sulphidic minerals such as pyrites

(FeS_2) in a mine, then release of the minewater into the surrounding water bodies (Ritcey, 1989). Furthermore, once the pyrites are exposed to water and oxygen, acidic waters containing metals and sulphates could be produced (Brown et al., 2002). It is well known that minerals are below the water level and at the anaerobic zones in natural conditions, where the oxidation by weather is very slow (Younger et al., 2002).

In detail, the first initiator chemical reaction is oxidisation of pyrites with releasing of hydrogen ions (H^+), which drops the pH of the water dramatically converting it to be acidic by sulphuric acid (Stumm and Morgan, 2012). The acid then solubilises other associated metals present in the rock, usually including aluminium and manganese. Ferrous (Fe^{2+}) and ferric iron (Fe^{3+}) is then produced by further oxidisation processes when contacting with dissolved oxygen of the water or atmosphere. Among other forms, ferric hydroxide ($Fe(OH)_3$) precipitates out of the water column as sludge, when pH is above 3.5, and that sludge is well known as ochre. Furthermore, microbiological processes can affect the oxidisation of ferrous iron to ferric iron. However, further acidity and ferrous iron could be formed with lowering the pH, reducing any remaining ferric iron. Acidic mine water drainage from disused mines has serious effects on the ecological system and chemical status of natural water by coating stream beds and aquatic plants (Younger et al., 2002). Thus, treatment of acidic mine water drainage is crucial for environmental protection.

2.10.2. Treatment of Acidic Minewater Drainage (AMD)

Typically, treatment of acidic minewater drainage could be achieved by either active or passive technologies. Both methods focus upon encouraging the oxidation of ferrous iron to ferric iron to the formation of ochre. An active treatment system is often applied by chemical neutralisation of acidic water with adjustment of pH using materials

such as lime, limestone, caustic or ammonia (Evangelou and Zhang, 1995). These alkaline materials are used to raise the pH of the AMD which then results in the precipitation of iron, aluminium and other metals as metal hydroxide sludge with high water content.

Furthermore, neutralisation could be achieved by adding of oxidising agents or flocculants (Heal et al., 2003). In such systems, high solid contents are produced with precipitated ochre with an iron content around 15% of the dry weight.

On the other side, passive treatment systems are more desirable because the oxidisation of acidic minewater drainage occurs naturally, and these account for about 80% of total treatment systems, such as constructed wetlands (Gazea et al., 1996). Those wetlands act as a buffer between the mine discharge and the aquatic environment (Brown et al., 2002). In contrast with active systems, passive technologies create very low solids contents associated with the produced ochre sludge (< 5%) with approximate iron contents of around 30% of ochre dry weight (Skousen, 1997). Therefore, ochre contains all of the contaminants that have precipitated out of solution, and any chemicals added to aid this process. The chemical composition of ochre thus varies with site and treatment method (Hedin, 2006).

Ochre sludge is normally generated in aquatic conditions with high moisture contents (80–90%). For easier handling, disposing and transporting, and reducing its weight as well, ochre is dried physically or chemically and often stored as stockpiles at treatment plant sites (Littler et al., 2013).

In the UK, the mine sites are managed by the Coal Authority which treat and consider all predominantly circumstances of the neutral ferruginous minewater with about 4500 tonnes of ochre (dry weight) per annum (Sapsford et al., 2015). Furthermore,

collection and disposal of this ochre waste sludge incurs a major operating cost for acidic mine drainage treatment facilities through storing within systems and/or dewatering and most is used for landfill because there is currently no established beneficial reuse for ochre sludge in the UK. Landfill disposal costs for ochre are in the range £79 per tonne to £129 per tonne at a median 50% solids (Sapsford et al., 2015).

All suggested possibilities for ochre sludge reuse and those under investigation consider consumption of the ochre for more beneficial and sustainable applications while minimising the transport cost in different fields. In the UK, ochre has been utilised in commercial application as a pigment in industry sectors (Hedin, 1998, 1999), as a filler in brick and cement manufacture (Dudeney, 1997), and as an iron source in the steel industry (Mayes et al., 2009). In the environmental field, ochre has been considered as an artificial soil additive (Evenson and Nairn, 2000); as a phosphorus adsorbent for soil amendment (Fenton et al., 2012); for phosphate removal in sewage treatment plants, in a natural sludge form (Wei et al., 2008); and in granular form as cement–ochre pellets (Heal et al., 2005; Sibrell et al., 2009; Littler et al., 2013).

2.10.3. Mechanisms of Phosphorus Removal by Ochre

Ochre sludge, with a majority of hydrous ferric oxide in its content, produced during treatment of acid mine drainage has been used for removal of phosphorus, in terms of phosphate ions, from contaminated wastewater. The mechanism of phosphorus removal onto the surface of goethite (α–FeOOH), a constituent of ochre sludge, is adsorption (Heal et al., 2003; Fenton et al., 2009). Whilst, Al– and Mg–hydroxides and calcium carbonates are also materials known for phosphorus adsorption (Heal et al., 2005).

Since phosphate ions have negative charge, ions with positive charge such as aluminium, iron, magnesium, and calcium will attract phosphate chemically. Therefore, mechanisms of phosphate removal depend on the chemical composition of ochre solids, and the level of pH in the aqueous solution is the control factor on the removal mechanisms.

Goethite is positively charged below pH values of 8.0–8.5 and generates increasing amounts of positive charge with declining pH. Therefore, increasing of charge and subsequent availability of surface sites will enhance the adsorption mechanism, with decreasing pH. However, with a high pH condition (> 7.0), phosphate ions are precipitated as calcium phosphate and/or adsorption onto calcite surfaces (Valsami–Jones, 2004). While, under low pH conditions (< 6.0) mechanisms of adsorption are dominant onto aluminium, iron, magnesium and clay surfaces.

In the pelletisation process, mixing a Portland cement binder with ochre sludge is leading to significant increases in the capacity of produced solids to adsorb phosphate compared with the raw ochre sludge. Furthermore, Littler et al., (2013) reported that cement–bound ochre continues to remove phosphorus for up to seven days while the equilibrium condition between raw ochre and phosphate solution appeared after approximately 60 minutes of contact time.

Removal of phosphorus by adsorption is a dominant mechanism in treatment with raw ochre sludge while precipitation of calcium phosphate was indicated when using cement–ochre pellets for phosphorus removal from artificial phosphate solution. Therefore, precipitation formulation generates an amorphous shape of particles with high level of supersaturation, presented generally as a fast process (Littler, 2012).

2.11. Recycling of Treated Wastewater

There is increasing interest in the reuse of wastewater in many countries around the world. Therefore, the driving force for reuse of wastewater is environmental and economic considerations. Wastewater reuse for non–drinking purposes offers a promising contribution to minimising the total costs of wastewater handling (Santos et al., 2014), since it contributes to reducing the load on wastewater treatment plants on the one hand, and on supplying water on the other.

There is interest in the possibility of reusing greywater because of its low level of microbial pollutants (Winward et al., 2008), mainly caused from human excreta, urine, bidets, and toilet paper, compared with municipal wastewater (Eriksson et al., 2002). Therefore, maximise recycling and/or reuse of the majority of domestic wastewater, such as greywater (GW), within households and rural small communities could also lead to control pollution (Houshia et al., 2012; Vakil et al., 2014).

Health aspects, environmental perspectives, and economic feasibility are vital characteristics for the evaluation of the potential reuse of greywater. These characteristics play a valuable role in the choice of direction of greywater reuse and/or the need for treatment and consequently the treatment technology (WHO, 2006b).

Concerning health aspects, spreading of diseases associated with reuse of untreated greywater can be a serious issue. Microorganisms in water, or their growth and breeding in a system, pose a risk of spreading through handling, contact, and/or aerosols (Christova–Boal, et al., 1996; Dixon et al., 1999; Ottoson and Stenström, 2003; Friedler et al., 2006).

Several countries and states are working on the guidelines for reuse of treated wastewater for non–potable options. In the USA, California has limited the levels to a

maximum of 2.2 of total coliforms per 100 ml in reclaimed water for use in toilet and urinal flushing, commercial laundries and in decorative fountains.

In Florida, reclaimed water for toilet flushing and for the irrigation of recreation areas must contain no detected faecal coliforms per 100 ml (Crook and Surampalli, 1996). World Health Organization guidelines for treated wastewater used for irrigation of agricultural crops and public sports fields limit faecal coliforms to < 1000 per 100 ml and *nematodes* to < 1 per litre (WHO, 1989).

In Australia, guideline values of thermotolerant coliforms are set on four levels, for recreational applications these are < 150 per 100 ml and for higher contacts e.g., irrigation of salad vegetables, they are lighter at < 10 per 100 ml (Gregory et al., 1996).

In Germany, the corresponding limits are *total coliforms* < 100 per ml and faecal coliforms < 10 per ml as well as *Pseudomonas aeruginosa* < 1 per ml (Nolde, 1999).

From an environmental perspective, soil may be destroyed when receiving untreated greywater through reusing for irrigation or infiltration due to accumulation of pollutants therein (Travis et al., 2010).

Many plants suffer due to their sensitivity to alkalinity of agricultural soil (Christova–Boal et al., 1996). Soaps are synthetic alkaline salts of long chains of fatty acids, while cleaning detergents are chemical products consisting of surfactants (WilliamáMcBain et al., 1912; Myers, 1988).

Due to the low levels of contaminating pathogens and nitrogen (Benami et al., 2015), reuse and recycling of greywater have gained particular interest (F. Li et al., 2009).

Indoor reuses of greywater are common practice in some countries, such as toilet flushing and for urinals, since they are often otherwise supplied with water of drinking quality (March et al., 2004). This indoor reuse could save around 30 percent of the total

water volume consumed domestically (Karpiscak et al., 1990). Nolde, (1999) has proven that greywater generated from bathrooms can meet the requirements of health and economy to reuse for flushing in a dual system. Surendran and Wheatley (1998) have recommended the recycling of greywater from washing machines or stormwater, as well as bathrooms, in multi–storey buildings for non–potable destinations.

The main outdoor applications of wastewater reuse, or greywater in particular, are essentially in agricultural sectors (Okun, 1997; Gross et al., 2014; Santos et al., 2014), such as lawns, athletic fields, cemeteries, parks and golf courses, as well as in home gardens.

For industrial processes and manufacturing, reuse of grey wastewater is also an option (Surendran and Wheatley, 1998). Washing vehicles, windows and streets, fire protection, boiler feed water and concrete production are examples of other suggested on–site usages (Pidou et al., 2008).

Furthermore, urban and environmental sectors could be developed when greywater is recycled, such as wetlands preservation (Otterpohl et al., 1999; Scholz, 2006). An alternative way of handling grey wastewater is to infiltrate it into the ground and thereby create a shortcut in the urban hydrological cycle (Eriksson et al., 2002).

Chapter Three

Materials and Methods

3.1. Overview

The research methodology is essentially focused on collection and analysis of the data from the experiments linked to the interactions between ranges of different design set–ups of operational variables and the effect of these interactions on the synthetic greywater treatment efficiency in floating treatment wetlands. In order to achieve these requirements, this chapter describes the following:

- Materials for the experiment: before starting the experiment, all materials were purchased such as, chemical materials of synthetic greywater, *P. australis* for floating treatment wetlands, plastic buckets, and chemical reagents for water quality tests.
- Greywater preparation: in order to operate the experimental work, artificial greywater was prepared to utilise as inflow to the floating treatment wetlands. Then, water samples of outflows were collected for analysis.
- Laboratory analysis: both physical and chemical water quality parameters for inflow and outflow discharges were measured at the laboratory. The parameters included colour, temperature, pH, redox potential, turbidity and total suspended solids, electrical conductivity, dissolved oxygen, chemical oxygen demand, 5–day biochemical oxygen demand, nutrients (phosphorus and nitrogen), and trace elements. Furthermore, materials and protocols were prepared for heavy metals and other elements in inflow and outflow greywater. The concentration of elements and the effect of greywater types were investigated by analysis for accumulation

of those elements into different parts of *P. australis* tissues and cement–ochre pellets.

- Data analysis: A large body of quantitative experimental data was collected, as expected, which required analysis by appropriate statistical methodologies

3.2. Synthetic Greywater

Household greywater was created artificially by using analytical–grade chemicals (Table 3.1) purchased from Fisher Scientific Co. Ltd. (Bishop Meadow Road, Loughborough, UK). The synthetic greywater was prepared under non–sterile conditions as a stock solution by mixing the selected chemicals with de–chlorinated public mains tap water at a temperature of around 25 °C. The resulting quality of greywater was simulated to mimic the qualities of real greywater (Tables 2.1–2.4) in terms of biochemical oxygen demand, chemical oxygen demand, ammonia–nitrogen, nitrate–nitrogen, ortho–phosphate–phosphorus, pH, redox potential, turbidity, total suspended solids, electrical conductivity, and trace elements as well. The resultant key pollutants of the proposed recipes are summarised in chapter four.

Furthermore, human hair has been assessed for its content in terms of metals as a function of different washing products (Chojnacka et al., 2012). Varying concentrations of metals can be linked to discharges of bath, shower and hand washing basins, while kitchen discharge is commonly less rich in metals (Kariuki et al., 2012). In general, phosphorus, sodium, iron, copper, cadmium, and chromium concentrations were highest in laundry greywater. The highest values of zinc and magnesium were found in kitchen greywater from households without sewers (Kariuki et al., 2012). Other unexpected sources of metals are linked to pharmaceutical products and rechargeable batteries.

Two stock solutions were mixed separately to represent low concentration (LC) and high concentration (HC) pollutant strengths of synthetic greywater strength (SGW). Chemical recipes were subsequently added to tap water (Figure 3.1) and stirred by a magnetic stirrer (3.0 cm long and 0.5 cm wide) with rounded edges for one hour at 1200 rpm (Schäfer et al., 2006). The concentrated stock solutions were diluted with tap water at a volume ratio of 1:100 before use in the experiment (Abed and Scholz, 2016). The two solutions were stored overnight at 4 °C, and stirred for a further 30 minutes before being used in the subsequent experiments (Nghiem, et al., 2006). The concentration levels of the proposed synthetic greywater are shown in Table 3.1. These concentrations were subject to environmental conditions typical for Greater Manchester (temperate and oceanic climate), after dilution of one litre of SGW's stock solution was with100 litres of tap water to fill the buckets.

Figure 3.1. Stages of preparing synthetic greywater (SGW).

Sodium hydroxide (NaOH) and hydrochloride acid (HCl) were used to adjust the pH value of the solution (Nghiem et al., 2006). A wide range of pH values for real greywater has been reported in literature (Table 2.1). However, in this experiment, the pH values for both low– and high– strength greywaters were adjusted at pH ranges of around 5–7 and 7–10, respectively (Abed and Scholz, 2016).

Table 3.1. Proposed ingredients for low– (LC) and high– (HC) contamination contents of synthetic greywater (SGW).

Item	Chemical name	Chemical formula	Molar mass (g/mol)	Low concentration (mg/l)	High concentration (mg/l)	Composition percentages
1	Kaolin	$Al_2Si_2O_5(OH)_4$	258.16	15	100	Al (20.90%), H (1.56%), O (55.78%) and Si (21.76%)
2	Cellulose	$(C_6H_{10}O_5)_n$	162.14	15	100	C (44.45%), H (6.22%) and O (49.34%)
3	Humic acid	$C_{187}H_{186}O_{89}N_9S_1$	4015.55	5	20	C (55.90%), H (4.67%), O (35.46%), N (4.67%) and S (0.80%)
4	Sodium chloride	NaCl	58.44	10	120	Cl (60.66%) and Na (39.34%)
5	Sodium hydrogen carbonate	$NaHCO_3$	84.01	10	85	C (14.30%), H (1.20%), Na (27.37%) and O (57.14%)
6	Calcium chloride	$CaCl_2$	147.02	10	55	Ca (36.11%) and Cl (63.89%)
7	Potassium nitrate	KNO_3	101.10	0	90	K (38.67%), N (13.85%) and O (47.48%)
8	Calcium nitrate	$Ca(NO_3)_2$	164.09	0	150	Ca (24.43%), N (17.07%) and O (58.50%)
9	Magnesium sulphate	$MgSO_4$	120.37	2	240	Mg (20.19%), S (26.64%) and O (53.17%)
10	Monopotassium phosphate	KH_2PO_4	136.09	13	85	H (1.48%), K (28.73%), O (47.03%) and P (22.76%)
11	Iron(III) chloride	$FeCl_3$	162.20	0.3	50.0	Fe (34.43%) and Cl (65.57%)
12	Boric acid	H_3BO_3	61.83	0.6	3.0	H (4.89%), B (17.48%) and O (77.63%)
13	Manganese(II) chloride	$MnCl_2$	125.84	0.03	3.20	Cl (56.34%) and Mn (43.66%)
14	Zinc sulphate	$ZnSO_4$	161.44	0.25	15.00	O (39.64%), S (19.86%) and Zn (40.50%)
15	Copper sulphate	$CuSO_4$	159.61	0.025	7.00	Cu (39.81%), O (40.10%) and S (20.09%)
16	Ammonium molybdate tetrahydrate	$(NH_4)_6Mo_7O_{24}$	1163.94	0.35	0.35	H (2.08%), Mo (57.71%), N (7.22%) and O (32.99%)
17	Cadmium oxide	CdO	128.41	0.02	12.50	Cd (87.54%) and O (12.46%)
18	Nickel oxide	NiO	74.69	0.02	0.06	Ni (78.58%) and O (21.42%)
19	Chromium(III) nitrate	CrN_3O_9	99.99	0.045	70.00	Cr (21.85%), N (17.65%) and O (60.50%)
20	Sodium sulphate	Na_2SO_4	142.04	2.60	25.00	Na (32.37%), O (45.06%) and S (22.57%)
21	Sodium phosphate monobasic	H_2NaPO_4	119.98	0.00	250.00	H (1.68%), Na (19.16%), O (53.34%) and P (25.82%)
22	Lead(II) oxide	Pb_3O_4	685.60	0.16	1.40	Pb (90.67%) and O (9.33%)
23	Secondary treatment effluent with microbial content (ml/l)	–	–	20.00	100.00	–

Note: Al = aluminium, H = hydrogen, O = oxygen, Si = silicon, C = , N = nitrogen, S = sulphur, Cl = chlorine, Na = sodium, Ca = calcium, K = potassium, Mg = magnesium, P = phosphorus, Fe = iron, B = boron, Mn = manganese, Zn = zinc, Cu = copper, Mo = molybdenum, Cd = Cadmium, Ni = nickel, Cr = chromium, Pb = lead, and item 23 was not considered in this study.

3.3. Cement–Ochre Solids

3.3.1. Deerplay Ochre Sludge

Ochre applied in this investigation originated from the mine wastewater treatment plant at the Deerplay coal mine site (53°44'06"N 2°11'49"W) north of Rochdale (OL13 8RD) in Lancashire, UK (Figure 3.2). The solids generated during the treatment of drainage from this deep coal mine were stockpiled in drying beds on–site. Ochre was collected in sealed ten–litre buckets, and transported to The University of Salford, where they were stored at room temperature within a laboratory. Raw ochre has a very high water content. Therefore, a 100–g sample of the sludge was taken for moisture content determination. This was defined through air drying of the sample in an oven at 105 °C for 24 hours. The moisture content was 87%.

Figure 3.2. Ochre sludge collected from Deerplay Minewater Treatment Site.

3.3.2. Pelletisation

In order to produce ochre pellets, ordinary Portland cement was added to the raw ochre sludge at a proportion of 30% by weight of the total blend. This percentage was similar to previous studies (Sibrell et al., 2009; Littler et al., 2013). The remaining 70% of the total blend weight was ochre sludge and water. It was desired to achieve 23% water in the mixture (Carr, 2012; Littler, 2012), but the moisture content of raw ochre sludge (87%) was higher than the target water to cement ratio (0.77), resulting in a compressive strength of around 20–25 MPa (Littler, 2012). Therefore, a large sample of raw ochre was dried at 40 °C within an oven for two days.

The dried ochre clods were crushed to fine powder. At 47% of the total blend weight, dry ochre was added to Portland cement, and mixed carefully for 15 minutes to obtain a homogenous consistency (Littler, 2012).

A dough concrete mixer model A200 HOBART (Southgate Way, Orton Southgate, Peterborough, UK) was set to its highest rate (361 rpm), and was frequently stopped to allow the material surrounding the vessel walls to be scraped off and returned to the mixer. Then, the mixture was transferred into 100×100×500 mm steel moulds, which were filled in three stages using a palette knife and compacted with a mechanical vibrator. Plastic foil was used to cover the cast to minimise moisture losses.

The material was left to cure for 28 days. Thereafter, the block was removed from the steel cast and dried within an oven at a temperature of 40 °C for two days (Figure 3.3). Thereafter, the block was well–crushed with a hammer into pellets, which were dry–sieved through a sieve with size ranges 3–5 mm, 5–10 mm and 10–20 mm on a vibrating separator.

Figure 3.3. Cement–ochre block before crushing.

For small batch experiments, particles with sizes less than 3 mm were used to investigate the characteristics of cement–ochre pellets, while pellets with a range of 3–5 mm were utilised to assess ochre, which was required for mesocosm–scale experiments simulating stabilisation ponds. Equal weights from the three size ranges of cement–ochre pellets were mixed and transferred into the pond treatment systems to evaluate their ability to treat two different contamination loads of synthetic greywater.

3.3.3. Determination of pH, Redox Potential and Electrical Conductivity of Ochre

According to ISO–10390 (ISO, 2005), raw ochre sludge and cement–ochre pellets were characterised according to pH, redox potential (Eh) and electrical conductivity (EC). Air–dried samples of particles that passed through a 3–mm sieve were crushed and subsequently passed through a 100–µm sieve; then 5 ml of crushed sample were inserted

into a centrifugal tube of 50–ml size. About 25 ml of deionised water were added to achieve a 1 : 5 volume ratio. Centrifuge cabinet shakers were operated for 60 minutes at a speed of 150 rpm.

Then the tube was left for one hour to allow for particles to settle down. The pH and Eh were measured with a sensION+ benchtop multi–parameter meter (Hach Lange, Düsseldorf, Germany), shown in Figure 3.4–a, while the electrical conductivity (EC) was measured by a conductivity meter entitled METTLER TOLEDO FIVE GOTM (Keison Products, Chelmsford, Essex, England, UK), shown in Figure 3.4–b.

Figure 3.4. Laboratory meters for; a) pH and redox potential (Eh) measurements, and b) electrical conductivity (EC) measurements.

3.3.4. Trace Element Analysis

Water and solid samples were collected and digested according to the USEPA (2014), SW–846:Test Method 6010D, for trace elements in waters and wastes to analyse for elements by Inductively Coupled Plasma–Optical Emission Spectrometry (ICP–OES) using a Varian 720–ES (Agilent Technologies UK Ltd., Wharfedale Road, Wokingham, Berkshire, UK), as shown in Figure 3.5.

Figure 3.5. Inductively Coupled Plasma–Optical Emission Spectrometry (ICP–OES) Varian 720–ES

According to USEPA (1994), Method 200.7, the chemical composition of the raw ochre sludge and the cement–ochre pellets were analysed before and after utilisation in the experiment. Acid digestion of solid samples was prepared according to USEPA (1996) Method 3050B. Specifically, 10 ml of aqua–regia were mixed with one part nitric acid (HNO_3) and three parts hydrochloric acid (HCl) were added to 250 mg of oven–dried solid samples, weighed on a digital balance at an accuracy of 0.1 mg. Then, samples were digested by a microwave accelerated reaction system type CEM Mars Xpress microwave (Figure 3.6). After digestion, samples were analysed for the trace elements aluminium (Al), boron (B), calcium (Ca), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), potassium (K), magnesium (Mg), manganese (Mn), sodium (Na), nickel (Ni) and zinc (Zn). Trace elements for each sample were recorded in mg/kg.

Figure 3.6. Accelerated reaction system (CEM Mars Xpress Microwave).

Furthermore, the potential release of minerals and trace elements from either raw ochre sludge or cement–ochre pellets into water was assessed. A plastic bottle containing 100 ml of deionised water and 100 mg of crushed solids was shaken for two hours by an orbit shaker at ambient laboratory temperature of around 25±2 °C. After shaking, solid particles were allowed to settle down for two hours, then 10 ml of water were withdrawn for filtration through a 0.45–µm cellulose filter paper into a sampling tube. From each type of solid, triplicate sub–samples of 10 ml each were acidified before analysing them for element content using ICP–OES (Sibrell et al., 2009).

Blank samples were prepared for each test containing the same reagents and proportions as those used for the processing of samples. This was required to identify possible contamination resulting from either the reagents (namely, acids) or equipment (including filters) used during sample processing. In addition, blanks were periodically

tested to ensure detection limits conformed to supposed values. Three standard calibration solutions were run from time to time between the samples to account for instrumental drifts.

3.3.5. Small Batch Experiment

Under controlled laboratory conditions, small batch experiments consisting of two groups of duplicate 300–ml glass jars were conducted. For the first group, jars were filled with HC–SGW, while for the second group, jars contained LC–SGW. Dry cement–ochre pellets with the particle range 3–5 mm were weighed on digital balances to obtain 2, 4, 6, 8 and 10 g for both HC–SGW and LC–SGW groups of jars, representing 10, 20, 30, 40 and 50 g of ochre pellets per litre of SGW solution. Jars were sealed and shaken once, then left for 15 minutes for contact. Thereafter, water samples were collected for routine analysis. Glass jars were cleaned, and ochre pellets were replaced by new fresh ones. The trials were repeated for 30 minutes, 60 minutes, 24 hours and 48 hours of contact time.

The test results showed that a higher ratio of ochre pellets per volume of SGW give higher removals of phosphorus characterised as PO_4–P. However, pH values increased to between 10 and 13, which is well above the threshold value of 9 for the reuse of treated wastewater in agriculture for irrigation purposes (FAO, 2003). Therefore, a quantity of 30 g of ochre pellets/l was selected to treat SGW in mesocosm–scale experiments to optimise phosphorus removal (80–90%) at pH values between 8 and 10.

3.4. Floating Treatment Wetlands (FTWs)

Experimental mesocosm–scale systems may simulate natural or constructed retention ponds or floating reedbed islands, which may provide an appealing alternative cheap approach to the more conventional subsurface flow, surface–flow constructed

wetlands, solving problems associated with water eutrophication in urban areas. In addition, there is no requirement for additional land area.

The experiment was operated under real weather conditions located on the open top floor of the Newton Building at The University of Salford (53°29'09.3"N and 2°16'24.8"W), Greater Manchester, UK. The experiment was conducted from 1st September 2014 to 1st November 2016. Water quality analysis started on 1st November 2014. Two months were allocated for biofilm development in the systems. The synthetic greywater used in this study had two chemical formulations for low and high pollutant concentrations. Fourteen–litre plastic buckets (purchased from B&Q, Manchester, UK) were filled with ten litres of SGW.

All plants with bare–rooted *Phragmites australis* (*Cav.*) *Trin. ex Steud.* (Common reed), formerly known as *Phragmites communes* (Norfolk reed) according to Scholz and Lee (2005), were provided by VESI Environmental Ltd. (Little Island, Co. Cork, Ireland), as shown in Figure 3.7. They had the same growth history and maturity and originated from the same batch. Plant rhizomes and roots were washed carefully to remove all attached soil before using them in the experiment. The bucket size was considered sufficient to grow *P. australis* roots and rhizomes, which were fully submerged in the water to allow them to penetrate as much as possible into the free water volume (depth of about 300 mm and a diameter of roughly 250 mm). The volume of roots and rhizomes was approximately one litre for all vegetated buckets (Figure 3.8).

On one side, to assess the ability of constructed ponds in the presence of cement–ochre pellets with a combination with *P. australis* in treatment of greywater under the effect of two different levels pollutant concentrations. On the other side, the experiment variables were selected to assess the ability of wetland macrophytes to treat synthetic

greywater (SGW) of two different pollutant loadings under short (two days) and long (seven days) contact (retention) times. Retention times of two and/or seven days are commonly maintained for biological treatment technologies such as constructed wetlands to purify various types of wastewater (Tanner et al., 1995; Ge et al., 2016). As well as for various treatment technologies such as stabilisation ponds and constructed wetlands for purification of various types of wastewater (Littler et al., 2013).

Figure 3.7. *Phragmites australis (Cav.) Trin. ex Steud.* (Common reed) purchased from VESI Environmental Ltd., Little Island, Co. Cork, Ireland.

Figure 3.8. Plastic bucket filled with ten litres of synthetic greywater for treatment by floating *P. australis*.

In general, there were 72 mesocosms in total. They were broken–down into three main groups of buckets. The first group was designed for two days of contact time. This group comprised eight sets of wetlands (T1, T2, T3, T4, T5, T6, T7 and T8), while, the second group of wetlands was for seven days of contact time with sets named as T9, T10, T11, T12, T13, T14, T15 and T16 (Table 3.2). Each set had four replicates marked as number of bucket followed by letter (a, b, c, and d) for the identification of the replicates. Ten litres of synthetic greywater with high contamination contents (HC–SGW) were treated in wetlands T1, T2, T3 and T4 for a 2–day contact time and in wetlands T9, T10, T11, and T12 for a 7–day contact time. Greywater with low contamination contents (LC–SGW) was treated in wetlands T5, T6, T7, and T8 for a 2–day contact time, and in wetlands T13, T14, T15 and T16 for a 7–day contact time. Wetlands (T1, T2, T5, T6, T9, T10, T13 and T14) were vegetated with *P. australis*, while, about 300 g of cement–ochre pellets were selected to treat 10 litres of SGW in wetlands T2, T4, T6, T8, T10, T12, T14 and T16 (Figure 3.9). Therefore, a combination of floating *P. australis* with cement–ochre pellets was used in wetlands T2, T6, T10 and T14, as a part of the experimental investigation, while, wetlands T3, T7, T11 and T15 were left with only synthetic greywaters (without vegetation and without cement–ochre pellets). The experimental set–up design includes four sets of bucket wetlands with two replicates that are essentially controls receiving ten litres of clean de–chlorinated clean tap water. The control sets were numbered as letter C followed by the bucket number (1, 2, 3 or 4) and a letter (a/b) to allow for the identification of the replicates. The control wetlands C1 (vegetated) and C2 (unvegetated) were designated for two days of contact time, while C3 (vegetated) and C4 (unvegetated) were subjected to seven days of contact time.

Figure 3.9. Mesocosm scale experiment of floating treatment wetlands.

Table 3.2. Operational parameters in experimental set–up design of different treatment systems.

Wetland labels	*Contact time*		*SGW*		TW	*Vegetation*		*Cement–ochre*	
	2–day	7–day	HC	LC		with	without	with	without
T1	✓		✓			✓			✓
T2	✓		✓			✓		✓	
T3	✓		✓				✓		✓
T4	✓		✓				✓	✓	
T5	✓			✓		✓			✓
T6	✓			✓		✓		✓	
T7	✓			✓			✓		✓
T8	✓			✓			✓	✓	
T9		✓	✓			✓			✓
T10		✓	✓			✓		✓	
T11		✓	✓				✓		✓
T12		✓	✓				✓	✓	
T13		✓		✓		✓			✓
T14		✓		✓		✓		✓	
T15		✓		✓			✓		✓
T16		✓		✓			✓	✓	
C1	✓				✓	✓			✓
C2	✓				✓		✓		✓
C3		✓			✓	✓			✓
C4		✓			✓		✓		✓

Note: T1 to T16, treatment systems with four replicates; C1 to C4, control wetlands with two replicates; SGW, synthetic greywater; HC, high pollutant load SGW; LC, low pollutant load SGW; TW, tap water.

After a specific contact time of treatment, the simulated greywaters in the treatment wetlands were completely replaced by a volume of ten litres of fresh SGW. For vegetated wetlands, the inflow was slowly filled to avoid disturbing the biofilm and microorganisms in the roots and rhizomes system of *P. australis* (Table 3.2 and Figure 3.10). Figure 3.11 shows the overview statistical experimental set–up design including the impact of four variables, named: existence of plant *P. australis*, presence of cement–ochre solids, treatment contact time, and pollutant concentrations of synthetic greywater.

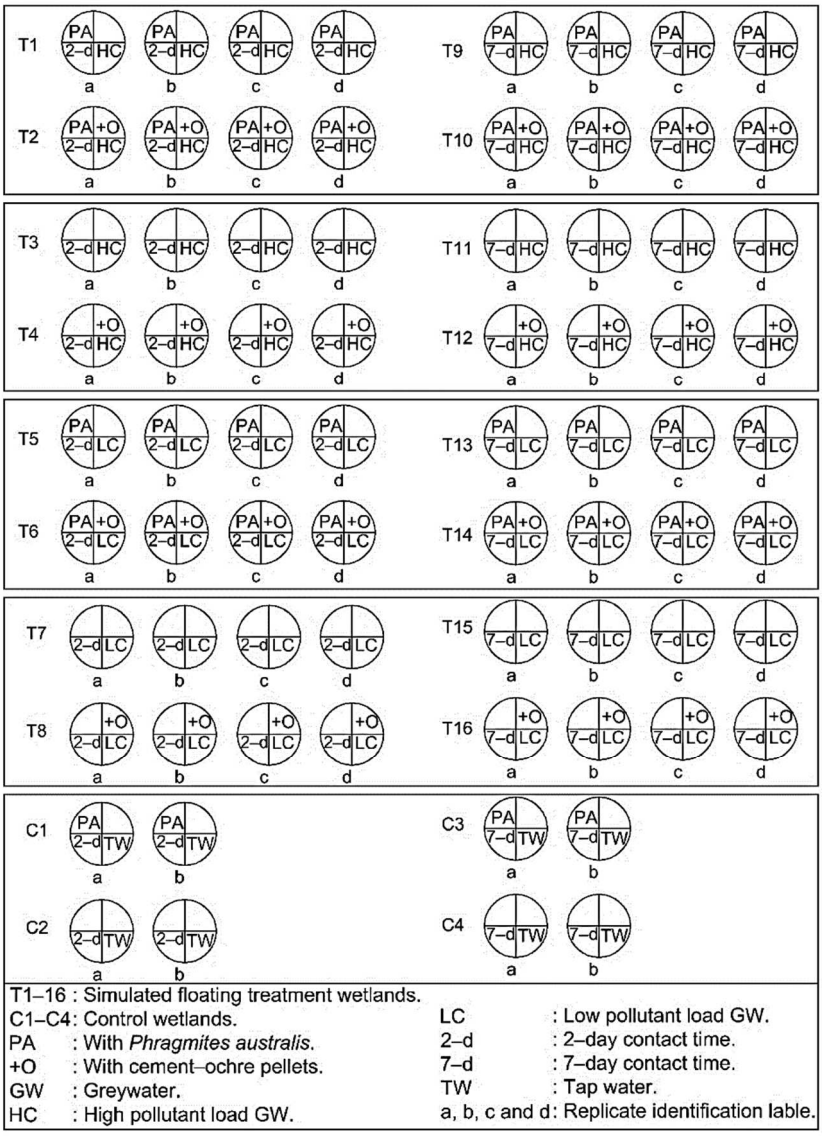


Figure 3.10. Experimental set–up design with different operational parameters in floating treatment wetlands.

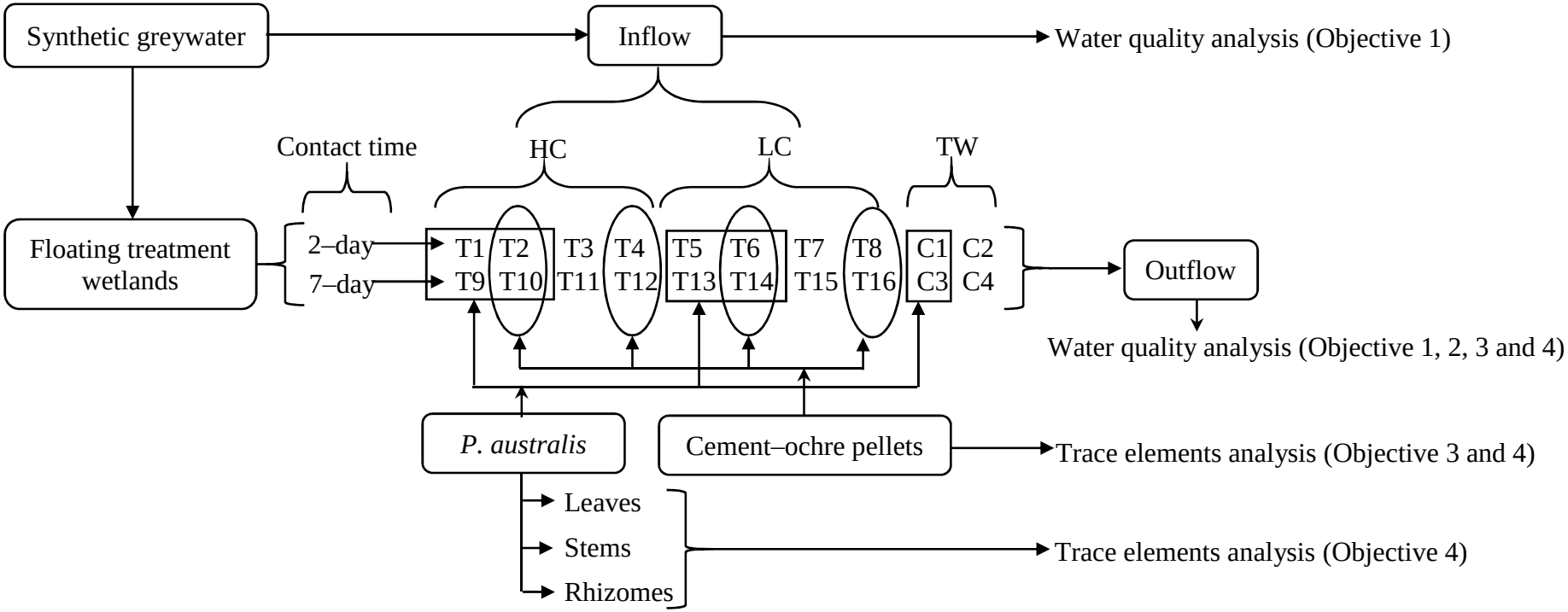


Figure 3.11. General layout of experimental methodology to achieve aim and objectives of the research.

3.5. Water Quality Analysis

Water quality sampling was performed according to APHA (2005), unless stated otherwise, to assess the properties of SGW. After the specific treatment time, 1000 ml of agitated outflow were collected for different water quality tests. All sampling containers were cleaned and washed with non–ionic detergent. They were subsequently rinsed with tap water, and later on soaked overnight in 10% nitric acid (HNO_3) solution. Finally, the containers were rinsed with deionised water before usage. In this study, the washing–out of solids and the possibility of them dissolving and/or being re–suspended back into the water column was considered. Furthermore, the focus of the study was on processes other than sedimentation, such as assessing the role of *P. australis* and adsorption capacity of cement–ochre pellets on the removal performances.

The spectrophotometer DR 2800 Hach Lange (www.hach.com) was applied for standard water quality analysis of variable parameters including chemical oxygen demand (COD), ammonia–nitrogen (NH_4–N), nitrate–nitrogen (NO_3–N), ortho–phosphate–phosphorus (PO_4–P), total suspended solids (TSS) and colour (Figure 3.12–a).

The 5–day biochemical oxygen demand (BOD_5) was measured for all water samples with the OxiTop IS 12–6 system, which is a mono–metric measurement device that was supplied by the Wissenschaftlich–Technische Werkstätten (WTW), Weilheim, Germany (Figure 3.12–b).

Turbidity was measured with a Turbicheck Turbidity Meter (Lovibond Water Testing, Tintometer Group), as shown in Figure 3.12–c.

Electrical conductivity (EC) was determined by the conductivity meter entitled METTLER TOLEDO FIVE GOTM (Keison Products, Chelmsford, Essex, England, UK)

(Figure 3.12–d). Hydrogen ion (pH) and redox potential (Eh) were measured with a sensION+ benchtop multi–parameter meter (Hach Lange, Düsseldorf, Germany) (Figure 3.12–e). Dissolved oxygen (DO) for all samples were measured by Digital Electrochemistry with an HQ30d Flexi Meter (Hach Lange, Düsseldorf, Germany), as shown in Figure 3.12–f.

Minerals and trace elements of inflow and outflow greywater were analysed following the SW–846: Test Method 6010D (USEPA, 2014), for trace elements in waters and wastes, by Inductively Coupled Plasma–Optical Emission Spectrometry (ICP–OES) using a Varian 720–ES (Agilent Technologies UK Ltd., Wharfedale Road, Wokingham, Berkshire, UK). Samples of 10 ml of greywater in plastic bottles were filtered through a 0.45–µm cellulose filter paper into a sampling tube. Triplicate samples of 10 ml from each treatment were acidified before analysing them for element content using ICP–OES (USEPA, 1994; Sibrell et al., 2009).

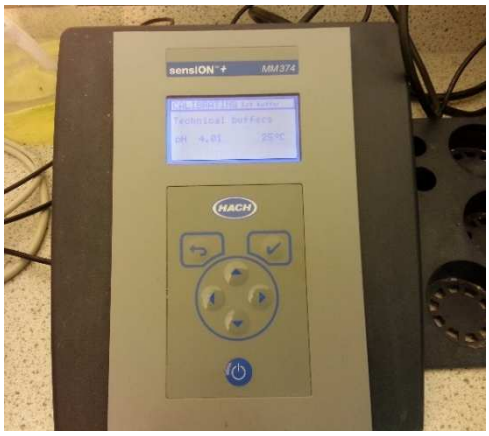
Blank samples were prepared for each test containing the same reagents and proportions as those used for the processing of samples. This was required to identify possible contamination resulting from either the reagents (namely, acids) or equipment (including filters) used during sample processing. In addition, blanks were periodically tested to ensure detection limits conformed to supposed values. Three standard calibration solutions were run from time to time between the samples to account for instrumental drifts.

a) Spectrophotometer DR 2800

b) OxiTop IS 12–6 system for BOD_5 measurement

c) Turbicheck turbidity meter

d) Electrical conductivity meter



e) SensION^{+} benchtop multi–parameter.

f) HQ30d Flexi dissolved oxygen meter.

Figure 3.12. Instrument meters used for water quality analysis.

3.6. Trace Elements Analysis

Trace elements were analysed for inflow and outflow synthetic greywater, and *P. australis* tissues (roots and rhizomes, stems, and leaves) for two growth seasons. Samples were collected and prepared for digestion according to specific analysis protocols for element indication by Inductively Coupled Plasma–Optical Emission Spectrometry (ICP–OES) using model Varian 720–ES (Agilent Technologies UK Ltd., Wharfedale Road, Wokingham, Berkshire, UK). According to USEPA (1994), water samples of 50 ml were preserved in glassware bottles at 4 °C. The samples were then acidified, if appropriate, by adding 1 ml of 70% concentrated nitric acid (HNO_3) to dissolve any suspended material in order to extract heavy metals and to reduce the pH to below 2.0, which was required for analysis. The samples were then filtered through a filter paper with a diameter of 0.45 µm prior to the analysis.

After finishing the experiment, *P. australis* tissue parts were carefully separated to dry overnight in an oven at a temperature of 105 °C to remove the water which was necessary for enzymatic reactions and to stabilise the sample (Plank, 1992). Specifically, 10 ml of aqua–regia were mixed with one part nitric acid (HNO_3) and three parts hydrochloric acid (HCl) were added to 300 mg of oven–dried plant tissue, weighed on a digital balance at an accuracy of 0.1 mg. Then, samples were digested in a CEM Mars Xpress microwave. After digestion, samples were analysed for the trace elements aluminium (Al), boron (B), calcium (Ca), cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), potassium (K), magnesium (Mg), manganese (Mn), sodium (Na), nickel (Ni) and zinc (Zn). Trace elements for each sample were recorded in mg/kg.

3.7. Statistical Analysis

Microsoft Excel was used for the general data analysis (e.g., mean, standard deviation, minimum and maximum values). In addition, Statistical Package for the Social Sciences (IBM–SPSS) Statistics Version 23 (www.ibm.com) was used to analyse all collected statistical data with 95% confidence level.

The independent samples T–test was used to compare sample means from two independent groups for an interval–scale variable when their distribution was approximately normal, while the non–parametric test (Mann–Whitney test) was used instead when the data were non–normally distributed. So, the hypothesis being tested was whether the two medians were equal (as opposed to two means in the independent samples T–test).

Therefore, it is important to examine the distribution of interval–scale data to check if they are normally distributed that is, bell–shaped, symmetrical about the mean. In addition to the histogram, normality tests such as Kolmogorov–Smirnov and Shapiro–Wilks can be used to decide if the distribution is normal. However, the Shapiro–Wilk test has been proved to have powerful results and be a more appropriate test for small sample sizes (< 50 tested samples) and can also deal with sample sizes as large as 2000 (Collis and Hussey, 2013). Therefore, a significance level (p–value) of the Shapiro–Wilk test is considered significant if it is less than 0.05 where the hypothesis of normality is rejected and the analysis must be operated by non–parametric techniques.

Furthermore, if there are more than two independent groups being compared in order variances analysis, the one–way ANOVA is used if the parametric assumptions are satisfied, that is if the interval–scale variable is approximately normally distributed (p–value of Shapiro–Wilk > 0.05). Otherwise, the non–parametric alternative is Kruskal–

Wallis one–way ANOVA and this is used for the interval–scale variables which are not normally distributed (p–value of Shapiro–Wilk < 0.05). Homogeneity of variances using Levene's test is necessary for both the parametric and non–parametric techniques.

For the parametric test, when the data meet the assumption of variances homogeneity, two possible tests could be used, either Tukey's honestly significant difference (HSD) or Scheffé post hoc tests. Otherwise, when the data do not meet the homogeneity of variances assumption, either the Games Howell or Dunnett's C post hoc test can be used. Moreover, the correlation relationships between variables were investigated using the Spearman's test since the data were non–normally distributed.

3.8. Strategy of Comparisons

Different strategies of statistical techniques were applied to compare the collected data in terms of inflow and outflow water quality and trace elements accumulated in the cement–ochre pellets and *P. australis* tissues.

Performances of treatment wetlands were compared with each other in order to distinguish the effect of operational variables in the experiment, such as impact of floating macrophytes, effect of cement–ochre pellet, effect of increasing contact time of treatment and the impact of pollutant concentrations of the inflow synthetic greywater. In chapter four, wetlands containing only synthetic greywater (T3, T7, T11, T15, C2 and C4) were considered in comparisons, to investigate the effect of storage time on greywater stability in terms of its quality. Wetlands with floating *P. australis* (T1, T5, T9, T13, C1 and C3) were considered in chapter five.

Those wetlands were compared with corresponding treatment systems without floating *P. australis*, and compared with each other in terms of contact time and inflow pollutant loads (Figure 3.13). In chapter six, wetlands treating the synthetic greywater

with only cement–ochre pellets (T4, T8, T12 and T16) were compared with corresponding systems which contained only greywater, and with each other for the effect of contact time and inflow pollutant loads (Figure 3.13). The effect of a combination between floating macrophytes and cement–ochre pellets were studied in chapter seven.

Wetlands T2, T6, T10 and T14 were compared with wetland systems treating greywater with only floating macrophytes (T1, T5, T9 and T13) and with wetlands (T4, T8, T12 and T16) which contained only cement–ochre pellets, respectively, to investigate the effect of those variables on treatment performances (Figure 3.13). In addition, wetlands with a combination system were compared with each other to determine the effect of contact time and inflow pollutant loads (Figure 3.13).

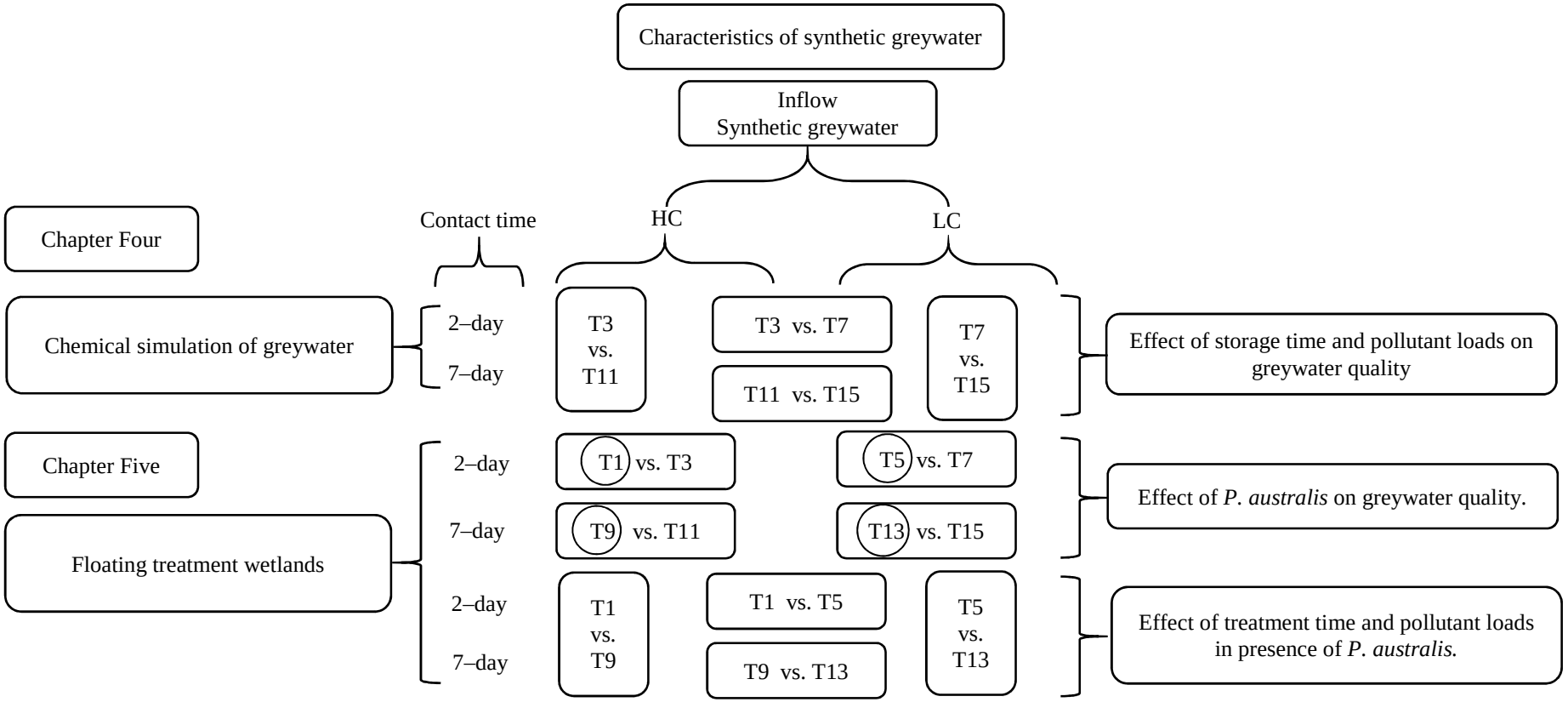


Figure 3.13. General layout of comparison strategy to achieve the research aim and objectives.

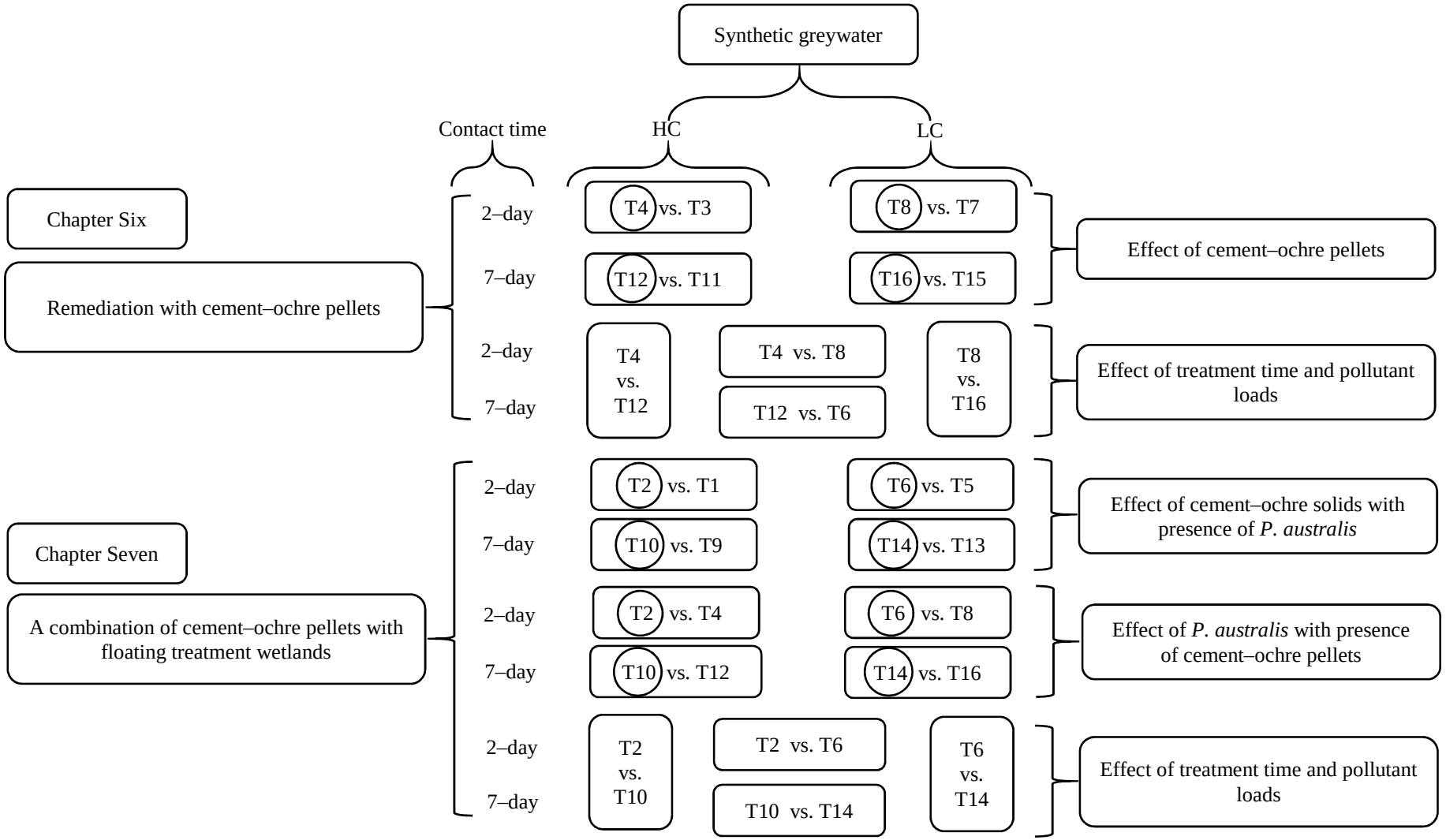


Figure 3.13. (Continued).

Chapter Four

Chemical Simulation of Greywater

Abed, S.N., Scholz, M., 2016. Chemical simulation of greywater. Environmental Technology, 37 (13), 1631–1646.

4.1. Overview

There is a need to develop standard synthetic greywater recipes to allow for the easy comparison of similar experiments in the future, to understand the removal mechanism for pollutants in greywater. Original experiments and a detailed literature review have been performed to support the development of reasonably stable generic synthetic greywater recipes for both low and high concentrations.

The scope of this chapter is limited to weak and strong standard synthetic greywater (SGW) recipe proposals being prepared under non–sterile conditions. It follows that specific greywater types, which are often a function of geographical region, and cultural and religious practices as well as guidelines and legislation, are beyond the scope of this article.

This chapter covers objective 1, section 1.5, through proposing practical recipes to be used for the simulation of greywater, which can be used with confidence to assess different treatment technologies. To achieve those objectives, stabilisation ponds were imitated, mentioned here as treatment systems named T3, T7, T11, and T15, which contained only synthetic greywater. These were considered to investigate the variation in greywater quality with time, since development of biofilm with associated microorganisms could change the water quality. In the experimental set–up design (section 3.4, Table 3.2, and Figure 3.10), wetlands T3 and T11 were operated to store high strength (HC–) synthetic greywater (SGW) for two and seven days, respectively.

While, wetlands T7 and T15 were operated to store low strength (LC–SGW) for two and seven days, respectively. The data were compared individually for inflow LC–SGW with their results of 2–day outflow and the same for 7–day outflow, alternatively, the same scenario was repeated for HC–SGW. The variation of water quality for the period between 2–day and 7–day storage time was also discussed.

4.2. Characteristics of Synthetic Greywater

The prepared synthetic greywater was used as inflow for the present experiment. The inflow water parameters in Table 4.1 refer to characteristics of prepared synthetic greywater, physiochemical parameters and trace element concentrations, just before utilisation in the experiment, for both LC and HC loads of pollutants.

Table 4.1. Physiochemical characteristics of inflow synthetic greywater (SGW).

Parameter	Unit	Inflow (low concentration)					Inflow (high concentration)				
		n	Mean	SD	Min	Max	n	Mean	SD	Min	Max
pH	–	81	6.9	0.48	5.3	7.9	81	8.4	1.61	5.4	11.5
Redox potential	mV	81	34.1	21.23	-18.1	111.2	81	-36.6	74.22	-182.1	97.9
Turbidity	NTU	81	22.9	7.14	9.8	41.6	81	188.9	47.22	18.3	308.0
Total suspended solids	mg/l	81	39.9	15.94	10.0	87.0	81	317.0	58.35	173.0	473.0
Electrical conductivity	µS/cm	81	164.6	63.24	98.7	452.0	81	988.5	196.09	612.0	1677.0
Dissolved oxygen	mg/l	81	10.4	1.24	7.7	12.3	81	10.5	1.39	6.9	12.6
Colour	Pa/Co	81	214.5	64.07	26.0	340.0	81	1587.8	379.89	787.0	2499.0
Temperature	°C	81	17.7	4.58	6.7	27.0	81	16.9	5.40	6.5	27.8
Biochemical oxygen demand	mg/l	81	17.6	8.00	2.0	40.0	81	34.7	12.99	10.0	60.0
Chemical oxygen demand	mg/l	81	28.9	14.47	8.2	86.7	81	129.2	34.68	63.9	221.0
Ammonia–nitrogen	mg/l	81	0.2	0.22	0.0	1.1	81	0.4	0.19	0.1	1.1
Nitrate–nitrogen	mg/l	81	1.3	1.21	0.1	7.6	81	8.9	6.38	0.2	29.8
Ortho–phosphate–phosphorus	mg/l	81	8.4	4.36	3.3	27.4	81	59.1	14.16	30.6	94.2
Element											
Aluminium	mg/l	45	0.52	0.528	0.09	1.56	45	2.13	0.869	0.76	4.77
Boron	mg/l	33	0.14	0.067	0.10	0.36	33	0.57	0.068	0.49	0.72
Calcium	mg/l	55	10.54	0.853	8.51	11.81	45	36.08	8.750	22.38	50.72
Cadmium	mg/l	42	0.09	0.056	0.04	0.23	39	7.36	2.981	3.67	11.58
Chromium	mg/l	54	0.04	0.063	0.01	0.23	54	3.20	0.918	1.67	5.70
Copper	mg/l	63	0.16	0.058	0.10	0.30	63	1.44	0.435	0.95	2.03
Iron	mg/l	48	0.21	0.102	0.11	0.43	48	6.41	2.476	2.48	10.37
Potassium	mg/l	12	4.04	0.448	3.41	4.65	12	60.16	1.684	58.31	62.53
Magnesium	mg/l	48	1.45	0.191	1.19	1.78	48	17.16	2.119	12.68	20.33
Manganese	mg/l	63	0.17	0.084	0.07	0.26	63	0.98	0.257	0.64	1.29
Sodium	mg/l	12	14.32	1.662	12.01	16.00	12	62.68	14.538	45.50	76.92
Nickel	mg/l	51	0.04	0.065	0.01	0.18	51	0.05	0.065	0.01	0.20
Zinc	mg/l	39	0.21	0.159	0.01	0.39	39	4.25	1.500	2.22	6.34

Note: n, number of tested samples; SD, standard deviation; Min, minimum; Max, maximum; and NTU, nephelometric turbidity unit.

In the literature, various reported recipes for synthetic greywater have been proposed to evaluate the efficiency of various types of treatment technologies, (Tables 2.5 and 2.6). This study illustrates how to choose analytical–grade chemicals to create two strength solutions of synthetic greywater (Table 3.1). In Table 4.1, organic and inorganic matter, dissolved and suspended solids, nutrients and macronutrients, trace elements and microorganisms were resembled carefully to simulate real greywater components and associated properties, (Tables 2.1–2.4).

These parameters were compared and discussed with published results of real greywater constituents obtained from previous research studies, (Tables 2.1–2.4). The figures shown in Table 4.1 are based on outdoor (greywater systems exposed to the elements) experiments.

4.2.1. The pH and Redox Potential

The ranges of pH and Eh for the inflow LC–SGW are comparable to a wide range of real greywaters whose values depend on the specific sources of greywater. There is a wide range of pH for real greywater and most of those were simulated by using LC synthetic greywater with a pH between 5.3 and 7.9, which linked to discharge from separated sources such as bathroom discharge (Christova–Boal et al., 1996) and from mixed sources of greywater (F. Li et al., 2009). While the pH values for HC greywater were simulated as values between 5.4 and 11.5, comparable to those real discharges which are commonly generated from laundries, as reported for real greywater (Christova–Boal et al., 1996; Eriksson et al., 2002; 2006; F. Li et al., 2009), and other SGW (Nghiem et al., 2006) in terms of pH. However, previous studies have not shown enough measurements related to Eh of real greywaters for a comparison to be made with those of the SGW used in this research

4.2.2. Turbidity and Total Suspended Solids

The reported ranges for turbidity and total suspended solids (TSS), as shown in Table 2.1, were successfully simulated, in particular by the ingredient of kaolin and cellulose fibres (Table 3.1) for both greywater strengths (Nghiem et al., 2006; Hourlier et al., 2010), as tabulated in Table 4.1. The simulated LC–SGW turbidity and TSS values (mean: 22.9, range: 9.8–41.6 and mean: 39.9 mg/l, range: 10.0–87.0 mg/l, respectively, Table 4.1) were linked to discharges from hand basins, showers and similar mixed greywater sources, as shown in Table 2.1, (Al–Jayyousi, 2003; Ramona et al., 2004; March et al., 2004; Pidou et al., 2008; Eriksson et al., 2010).

For HC–SGW, turbidity and TSS concentrations (mean: 188.9, range: 18.3–308.0 and mean: 317.0 mg/l, range:173.0–473.0 mg/l, respectively, Table 4.1) were similar to the characteristics of particles usually associated with the effluents from laundries, kitchens and mixed greywater sources (Eriksson et al., 2002; Al–Hamaiedeh and Bino, 2010).

4.2.3. Electrical Conductivity

High levels of electrical conductivity (EC) have been reported for previous references regarding discharges generated from laundries and some mixed real greywater sources (Eriksson et al., 2002; Christova–Boal et al., 1996). Therefore, the higher limits of EC were simulated by HC artificial greywater with a range between 612.0 and 1677.0 µS/cm. In contrast, low values of electrical conductivity are linked to bathroom fluxes and hand wash basins which were mimicked artificially between 98.7–452.0 µS/cm (Christova–Boal et al., 1996; Al–Hamaiedeh and Bino, 2010; Leal et al., 2012).

4.2.4. Dissolved Oxygen

The dissolved oxygen (DO) ranges for both LC and HC greywaters were also represented to mimic those reported, approximately between 9.6 and 12.6 mg/l, (Table 4.1). These were higher than those reported by Shin et al. (1998) and Eriksson et al. (2002), especially in the absence of sufficient numbers of microorganisms in the prepared artificial greywater, however, they were around the upper limits of published values (Leal et al., 2011).

4.2.5. Colour and Temperature

The test results of synthetic greywater have shown ranges of colour from 26.0 to 340.0 Pa/Co and from 787.0 to 2499.0 Pa/Co for low (LC) and high (HC) greywater concentrations, respectively, although there are very few reported data regarding the colour of real greywater. The temperature range was around 6.5–27.8 °C for both types of proposed greywater, which was similar to figures reported by Eriksson et al. (2002) and Christova–Boal et al. (1996).

4.2.6. Oxygen Demands

Almost all water quality parameters of the proposed synthetic greywaters, (Table 4.1), have similar values in terms of averages, or are at least within the published ranges (Tables 2.1–2.4). Although the concentrations of the 5–day biochemical oxygen demand (BOD_5), in particular for low–strength synthetic greywater (mean of 17.6 mg/l), are less than the reported values for real greywater, but they agree with those indicated by Winward et al. (2008) and Eriksson et al. (2010). Furthermore, the majority of published values of BOD_5 were approximately represented in the form of the HC synthetic greywater with a minimum value between 10.0 and 60.0 mg/l and mean around 37.8 mg/l, (Table 4.1).

The review on literature reveals that there is a wide variation in chemical oxygen demand (COD) concentrations of various greywater types and their compositions (Table 2.1). This can be explained by the great variety of household chemicals used causing a high degree of fluctuation from sample to sample (Eriksson et al., 2002; Al–Jayyousi, 2003; Eriksson et al., 2010). Comparing the analysis of synthetic greywater (Table 4.1), the COD concentrations of LC synthetic greywater range between 8.2–86.7 mg/l, and were similar to the lower limits of reported studies (Eriksson et al., 2002; Ramona et al., 2004; Pidou et al., 2008; Eriksson et al., 2010), while for HC greywater they were 63.9–221.0 mg/l with a mean value around 129.2 mg/l.

4.2.7. Biodegradability of Greywater

The biodegradability of greywater can be measured by the ratio between BOD_5 and COD. Previous research on pilot–scale biological systems treating greywater has shown that such technologies are generally limited to a low organic load, while analysis of the inflow greywater has shown a low BOD_5 : COD ratio associated with a lack of nitrogen and phosphorus (Laine et al., 1999). The addition of macro– and trace nutrients may stimulate biological processes and improve treatment efficiency (Jefferson et al., 2001). In this study, the BOD_5 to COD ratio of inflow SGW before utilisation in the experiment was 0.27 and 0.61 for HC–SGW and LC–SGW, respectively. Similarly, low biodegradability of greywater has been reported by Jefferson et al. (1999). However, after two hours of treatment, the BOD_5 to COD ratio of the greywater increased to 0.41, which is comparable with municipal wastewater, where the ratio is often between 0.4 and 0.8 (Marco et al., 1997).

In general, The BOD_5 and COD means were lower than those published for low–strength greywater linked to separated greywater effluents generated from hand basins,

showers and bath waters (Jefferson et al., 2004; Friedler et al., 2005). Although the concentrations of BOD_5 and COD in LC–SGW are less than some of the reported values for real greywater, they agree with those reported by Avery et al. (2007) and Winward et al. (2008). However, HC–SGW had a wide range of oxygen demand values due to the great variety of household products causing a high degree of instability from sample to sample (Eriksson et al., 2002; Al–Jayyousi, 2003).

4.2.8. Nutrients

Furthermore, the test results for synthetic greywater (Table 4.1) have shown appropriate simulations for reported values of ammonia–nitrogen (NH_4–N), nitrate–nitrogen (NO_3–N) and ortho–phosphate–phosphorus (PO_4–P), which were 0.0–1.1 mg/l, 0.1–7.6 mg/l, and 3.3–27.4mg/l, respectively, for LC synthetic greywater and 0.1–1.1 mg/l, 0.2–29.8 mg/l, and 30.6–94.2 mg/l, respectively, for HC synthetic greywater, in terms of mixed greywater, regardless of the sources of origin.

For biological wastewater degradation, a COD : N : P ratio of 100 : 20 : 1 has been suggested by Metcalf et al. (1991) and a ratio of 100 : 10 : 1 has been proposed by Beardsley and Coffey (1985). The HC–SGW and LC–SGW inflows were deficient and unbalanced in terms of nutrients and organic matter due to the exclusion of urine and excreta (Abed and Scholz, 2016). An optimised nitrogen to phosphorus ratio can make a significant difference to the rates of oxygen uptake and COD removal for both real and synthetic greywater (Al–Jayyousi, 2003).

Researches have indicated that greywater is nutrient–deficient and that the addition of nutrients has a positive effect on the biological treatment efficiency (Jefferson et al., 2001; 2004). Furthermore, NH_4–N levels in real greywater were significantly ($p < 0.05$) higher than those found in drinking water (Eriksson et al., 2002). A comparison

between the reported values for nutrients such as of NH_4–N, NO_3–N and PO_4–P in real greywater with those obtained from this study indicates similarities, regardless of the source of greywater (Schäfer et al., 2006; Diaper et al. 2008; F. Li et al., 2009; BS–8525–1, 2010; BS–8525–2, 2011; Hourlier et al., 2010). In general, the nitrogen constituents in greywater are lower than those in domestic wastewater; greywater contains only about a tenth of the nitrogen (ammonia, nitrite and nitrate) in domestic wastewater, since nitrogen is a major component of urine (Oron et al., 2014). In contrast, phosphorus is dominant in washing discharge wastes associated with detergents (Al–Jayyousi, 2003; Gross et al., 2007a; Al–Hamaiedeh and Bino, 2010).

In this study, the inflow LC–SGW showed an adequate biodegradation rate of 0.61 (in terms of the BOD_5 : COD ratio), which is similar to findings reported by F. Li et al. (2009). In comparison, the ratio was 0.27 for the inflow of HC–SGW.

4.2.9. Trace Elements

According to data shown in Tables 2.3 and 2.4, which are liked to trace elements and heavy metals in particular, synthetic greywater solutions represent reality reasonably well. The recipes were based on the molar weight of the chemical compositions multiplied by the percentage of the specific element in that chemical. For example, 100 mg of iron(III) chloride, item number 11, provides approximately 34 mg/l of iron, as illustrated in Table 3.1.

The reason for the appearance of metals such as these in greywater is that the personal care and cosmetic products which are used daily contain a wide range of trace elements from manufacture of pigments and other raw materials (Bocca et al., 2014). Furthermore, human hair has been investigated and found to have a heavy metal content due to utilisation of different local washing products (Chojnacka et al., 2012).

So, varying levels of heavy metal concentrations could be linked to discharges from baths, showers, and hand washing basins, while kitchen discharges could have a lower heavy metal content (Kariuki et al., 2012). In general, P, Na, Fe, Cu, Cd and Cr concentrations were highest in laundry greywater, and the highest values of Zn and Mg were indicated in greywater from non–sewered kitchen households (Kariuki et al., 2012). Other unexpected sources of metals might be through release of chemicals to greywater from sources such as pharmaceutical products and rechargeable batteries.

However, internal chemical reactions between different compositions, pH value, and cation ion interchangers have major effects on availability of dissolved elements for biological processes. Therefore, an Inductively Coupled Plasma–Optical Emission Spectrometry (ICP–OES) instrument was operated to analyse the inflow and outflow synthetic greywater samples to detect and identify the trace metal concentrations, Table 4.1.

4.3. Outflow Greywater Quality

Table 4.2 shows the water quality results of outflow HC and LC synthetic greywaters after two days (treatment systems T3 and T7, respectively) and seven days (treatment systems T11 and T15, respectively) of storage under real weather conditions (Figure 3.11).

Pollutant concentrations of outflow greywater after specific times of storages were compared with those concentrations before storage, to evaluate the overall stability of proposed synthetic greywater (objective 1, section 1.5 and Figure 3.13).

Table 4.2. Outflow water quality parameters for 2–day and 7–day contact times for treatment systems containing high (HC) and low (LC) contaminations of synthetic greywater (SGW).

Parameter	Unit	2–day outflow (HC)–T3						2–day outflow (LC)–T7					
		n	Mean	SD	Min	Max	Rem (%)	n	Mean	SD	Min	Max	Rem (%)
pH	–	85	7.8	1.37	5.6	9.8	na	85	7.5	0.70	6.3	10.1	na
Redox potential	mV	85	-3.0	62.95	-107.6	88.6	na	85	4.2	30.40	-116.1	51.0	na
Turbidity	NTU	85	192.1	50.87	102.0	341.0	-1.7	85	20.2	14.20	2.9	129.0	11.8
Total suspended solids	mg/l	85	321.8	56.68	165.0	447.0	-1.5	85	30.0	12.12	11.0	76.0	24.8
Electrical conductivity	µS/cm	85	965.2	106.68	627.0	1208.0	na	85	138.5	23.26	79.0	215.0	na
Dissolved oxygen	mg/l	85	10.2	0.73	8.7	12.1	2.9	85	10.5	0.82	8.2	12.6	-1.0
Colour	Pa/Co	85	1527.6	326.28	677.0	2311.0	3.8	85	164.5	40.93	34.0	265.0	23.3
Temperature	°C	85	17.1	4.75	6.1	23.6	na	85	16.0	4.59	5.3	21.8	na
Biochemical oxygen demand	mg/l	85	14.7	7.78	0.0	40.0	57.6	85	5.6	3.60	0.0	20.0	68.2
Chemical oxygen demand	mg/l	85	106.6	22.68	43.3	164.0	17.5	85	26.8	6.18	15.4	41.9	7.3
Ammonia–nitrogen	mg/l	85	0.4	0.16	0.0	0.9	0.0	85	0.09	0.05	0.0	0.3	55.0
Nitrate–nitrogen	mg/l	85	9.4	4.67	0.5	24.0	-5.6	85	1.2	0.71	0.1	3.2	7.7
Ortho–phosphate–phosphorus	mg/l	85	46.2	10.74	23.7	70.1	21.8	85	7.0	3.89	3.0	18.8	16.7
		7–day outflow (HC)–T11						7–day outflow (LC)–T15					
pH	–	83	7.7	1.21	5.9	9.9	na	83	7.5	0.72	6.4	9.3	na
Redox potential	mV	83	-4.4	59.67	-108.3	78.1	na	83	1.8	33.00	-87.9	53.2	na
Turbidity	NTU	83	185.7	49.24	65.1	281.0	1.7	83	16.5	7.27	5.7	34.1	27.9
Total suspended solids	mg/l	83	302.6	61.44	147.0	434.0	4.5	83	25.0	10.96	7.0	56.0	37.3
Electrical conductivity	µS/cm	83	1003.0	306.88	492.0	2460.0	na	83	144.0	32.28	97.7	263.0	na
Dissolved oxygen	mg/l	83	10.5	0.91	7.9	12.0	0.0	83	11.0	1.11	8.1	14.3	-5.8
Colour	Pa/Co	83	1644.8	489.96	718.0	2889.0	-3.6	83	152.6	41.05	51.0	258.0	28.9
Temperature	°C	83	16.6	3.87	7.6	22.1	na	83	15.3	4.23	6.7	22.2	na
Biochemical oxygen demand	mg/l	83	16.6	7.07	4.0	38.0	52.2	83	6.7	4.85	0.0	22.0	61.9
Chemical oxygen demand	mg/l	83	100.8	27.65	11.6	159.5	22.0	83	17.2	6.95	6.0	36.7	40.5
Ammonia–nitrogen	mg/l	83	0.3	0.13	0.0	0.8	25.0	83	0.1	0.04	0.0	0.3	50.0
Nitrate–nitrogen	mg/l	83	8.5	8.42	0.4	34.5	4.5	83	1.0	0.64	0.0	4.0	23.1
Ortho–phosphate–phosphorus	mg/l	83	43.0	13.78	20.25	79.4	27.2	83	8.5	4.03	2.6	19.6	-10.0

Note: n, number of tested samples; SD, standard deviation; Min, minimum; Max, maximum; Rem, removal; NTU, nephelometric turbidity unit; and na, not applicable.

4.3.1. Oxygen Demands

Inflow quality parameters were statistically compared with corresponding values of outflow after storage in treatment systems for two and seven days of contact time.

In terms of biochemical oxygen demand, although a standard nitrification inhibitor was utilised in determining BOD_5 (APHA, 2005), considerable BOD_5 outflow fluctuations may indicate the presence of nitrification processes during the incubator period (Tanner et al., 1995). Burgoon et al. (1989) reported that BOD_5 removal for unplanted mesocosms has been lower by around 15% when compared with vegetated systems (Figure 4.1–a and b).

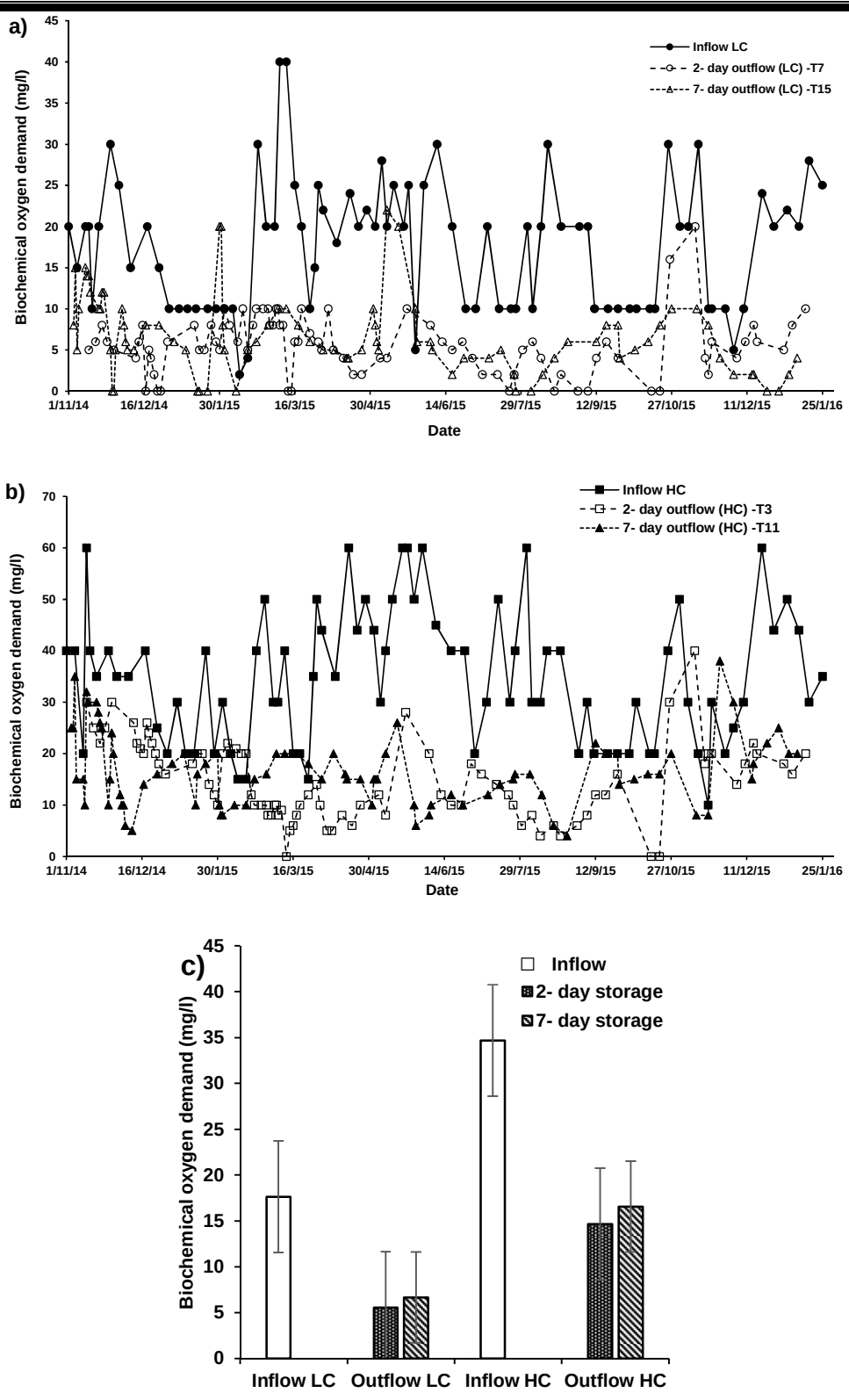


Figure 4.1. Effect of storage time on the variation of 5–day biochemical oxygen demand for synthetic greywater in: a) low concentrations (LC), b) high concentrations (HC), and c) the average values for low and high concentrations.

Table 4.3 shows that the outflow BOD_5 concentrations were significantly lower than those for both inflow greywater load strengths (HC–SGW and LC–SGW) in all treatment systems (Tables 4.1, 4.2 and Figure 4.1–c).

The variations in the BOD_5 concentrations of LC greywater (Figure 4.1–a) have shown significant ($p < 0.05$) reductions in the averages from 17.6 mg/l of the inflow to 5.6 mg/l and to 6.7 mg/l after two and seven days of storage time, respectively (Table 4.3, Figure 4.1–c). However, the BOD_5 concentration pattern (Figure 4.1–b) for HC greywater dropped significantly ($p < 0.05$) from 34.7 mg/l of inflow to 14.7 mg/l after two days of storage with a reduction rate of 57.6 %, and it was stable at around 16.6 mg/l for outflow water after seven days with a reduction rate of 52.2 % (Table 4.3 and Figure 4.1–c). This change has been confirmed by comparing available data evidence, which was reported by Jefferson et al. (1999), since, the microbial contamination is the likely reason for the drop in organic strength (Friedler, et al., 2006; Maiga, et al., 2014). The data variability is therefore high, resulting in some unexpected findings, which are, however, not statistically significant ($p > 0.05$).

However, there was no significant ($p > 0.05$) effect of increasing treatment time on BOD_5 concentrations in systems containing HC–SGW (T3 compared to T11) as shown in Table 4.4. The same trend regarding treatment time effects has been demonstrated for LC–SGW T7 with T15 (Figure 4.1–c). In this context, Tanner et al. (1995) argued that with increasing retention time of treatment in constructed wetlands, removals of BOD_5 increased by a rate of 16% and 25% for planted and unplanted wetlands, respectively. However, there was no significant ($p > 0.05$) effect of increasing treatment time on BOD_5 concentrations in control systems receiving tap water (C2 compared to C4).

Table 4.3. Significant values of the statistical analysis concerning the comparison between the inflow and outflow greywater after storage for two and seven days of contact time for both types of synthetic greywater (SGW).

		2–day contact time						7–day contact time					
		Inflow & T3 (HC–SGW)			*Inflow & T7 (LC–SGW)*			*Inflow & T11 (HC–SGW)*			*Inflow & T15 (LC–SGW)*		
Parameter	**Unit**	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
pH	–	<0.001	M–W	0.027	0.132	T–test	<0.001	<0.001	M–W	0.004	0.005	M–W	<0.001
Redox potential	mV	<0.001	M–W	0.004	0.013	M–W	<0.001	<0.001	M–W	0.004	<0.001	M–W	<0.001
Turbidity	NTU	0.118	T–test	0.676	<0.001	M–W	0.004	0.471	T–test	0.667	0.008	M–W	<0.001
Total suspended solids	mg/l	0.217	T–test	0.593	<0.001	M–W	<0.001	0.318	T–test	0.127	<0.001	M–W	<0.001
Electrical conductivity	µS/cm	0.012	M–W	0.376	<0.001	M–W	0.001	0.042	M–W	0.320	<0.001	M–W	0.013
Dissolved oxygen	mg/l	0.005	M–W	0.002	<0.001	M–W	0.846	0.004	M–W	0.301	<0.001	M–W	0.005
Colour	Pa/Co	0.218	T–test	0.274	0.483	T–test	<0.001	0.285	T–test	0.405	0.245	T–test	<0.001
Temperature	°C	0.005	M–W	0.923	0.015	M–W	0.023	0.004	M–W	0.585	0.002	M–W	0.001
Biochemical oxygen demand	mg/l	0.002	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Chemical oxygen demand	mg/l	0.005	M–W	<0.001	<0.001	M–W	0.811	0.002	M–W	<0.001	<0.001	M–W	<0.001
Ammonia–nitrogen	mg/l	<0.001	M–W	0.860	<0.001	M–W	<0.001	<0.001	M–W	0.091	<0.001	M–W	<0.001
Nitrate–nitrogen	mg/l	0.005	M–W	0.325	<0.001	M–W	0.687	<0.001	M–W	0.204	<0.001	M–W	0.201
Ortho–phosphate–phosphorus	mg/l	0.107	T–test	<0.001	<0.001	M–W	0.003	0.195	T–test	<0.001	<0.001	M–W	0.606

Note: HC, high concentration; LC, low concentration; SGW, synthetic greywater; p value, significant difference if $p < 0.05$ and not significant if $p > 0.05$; M–W, Mann–Whitney U–test; and NTU, nephelometric turbidity unit.

Table 4.4. Significant values of the statistical analysis for outflow greywater after storage for two and seven days of contact time to investigate the effects of increasing contact time and pollutant loads on changing values of parameters.

		Effects of increasing contact time						Effects of pollutant loads					
		HC–SGW			*LC–SGW*			*2–day contact time*			*7–day contact time*		
		T3 & T11			T7 & T15			T3 & T7			T11 & T15		
Parameter	**Unit**	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
pH	–	<0.001	M–W	0.751	0.003	M–W	0.805	–	–	–	–	–	–
Redox potential	mV	<0.001	M–W	0.732	0.002	M–W	0.797	–	–	–	–	–	–
Turbidity	NTU	0.318	T–test	0.405	<0.001	M–W	0.030	<0.001	M–W	0.006	<0.001	M–W	<0.001
Total suspended solids	mg/l	0.340	T–test	0.037	<0.001	M–W	0.009	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Electrical conductivity	µS/cm	0.002	M–W	0.492	0.275	T–test	0.197	–	–	–	–	–	–
Dissolved oxygen	mg/l	0.008	M–W	0.012	<0.001	M–W	<0.001	<0.001	M–W	0.069	0.655	T–test	0.047
Colour	Pa/Co	0.513	T–test	0.071	0.177	T–test	0.063	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Temperature	°C	<0.001	M–W	0.099	<0.001	M–W	0.112	–	–	–	–	–	–
Biochemical oxygen demand	mg/l	0.016	M–W	0.122	<0.001	M–W	0.264	<0.001	M–W	0.010	<0.001	M–W	0.031
Chemical oxygen demand	mg/l	<0.001	M–W	0.214	0.024	M–W	<0.001	0.018	M–W	0.032	0.001	M–W	<0.001
Ammonia–nitrogen	mg/l	0.029	M–W	0.033	0.107	T–test	0.303	<0.001	M–W	0.001	<0.001	M–W	<0.001
Nitrate–nitrogen	mg/l	0.001	M–W	0.005	<0.001	M–W	0.028	<0.001	M–W	0.826	<0.001	M–W	0.524
Ortho–phosphate–phosphorus	mg/l	0.001	M–W	0.025	<0.001	M–W	0.001	<0.001	M–W	0.664	<0.001	M–W	0.001

Note: HC, high concentration; LC, low concentration; SGW, synthetic greywater; p value, significantly different if $p < 0.05$ and not significantly different if $p > 0.05$; M–W, Mann–Whitney U–test; and NTU, nephelometric turbidity unit.

Furthermore, the investigation showed that BOD_5 decreased significantly in systems storing LC–SGW (T7: 2–day contact time and T15: 7–day contact time) compared to those systems of HC–SGW (T3: 2–day contact time, and T11: 7–day contact time) respectively, as shown in Table 4.4.

A comparison of the inflow COD value of HC–SGW with those of the outflows showed significant decreases in systems for both treatment contact times. Where the COD dropped from 129.2 mg/l in the inflow to 106.6 mg/l (reduction of 17.5%) and to 100.8 mg/l (reduction of 22.0%) for 2–day and 7–day storage times, respectively. Furthermore, the corresponding standard deviations are relatively high and the sample numbers of both data sets are different. Some of the COD data variations can be attributed to both experimental variability and biodegradation of the fraction of the COD which is biodegradable (Essington, 2004; Friedler et al., 2005). In contrast, the COD concentrations of the LC–SGW outflow decreased from 28.9 mg/l in the inflow to 26.8 mg/l, not statistically significant ($p > 0.05$), for the 2–day storage time experiment (system T7), while it dropped significantly to 17.2 mg/l for the 7–day storage time experiment (system T15), compared to the LC–SGW inflow values (Figure 4.2).

In addition, increasing the contact time led to a significant effect in reduction of the COD concentrations in the LC–SGW system (T15) compared to the outflow COD of system T7 (2–day contact time). However, there was no significant change between outflow COD values of systems treating HC–SGW when increasing the contact time of treatment (Tables 4.2 and 4.4).

For all systems, the COD removals from HC–SGW (T3 and T11) were significantly higher than those removals for systems treating LC–SGW (T7 and T15) for both treatment contact times (Table 4.4).

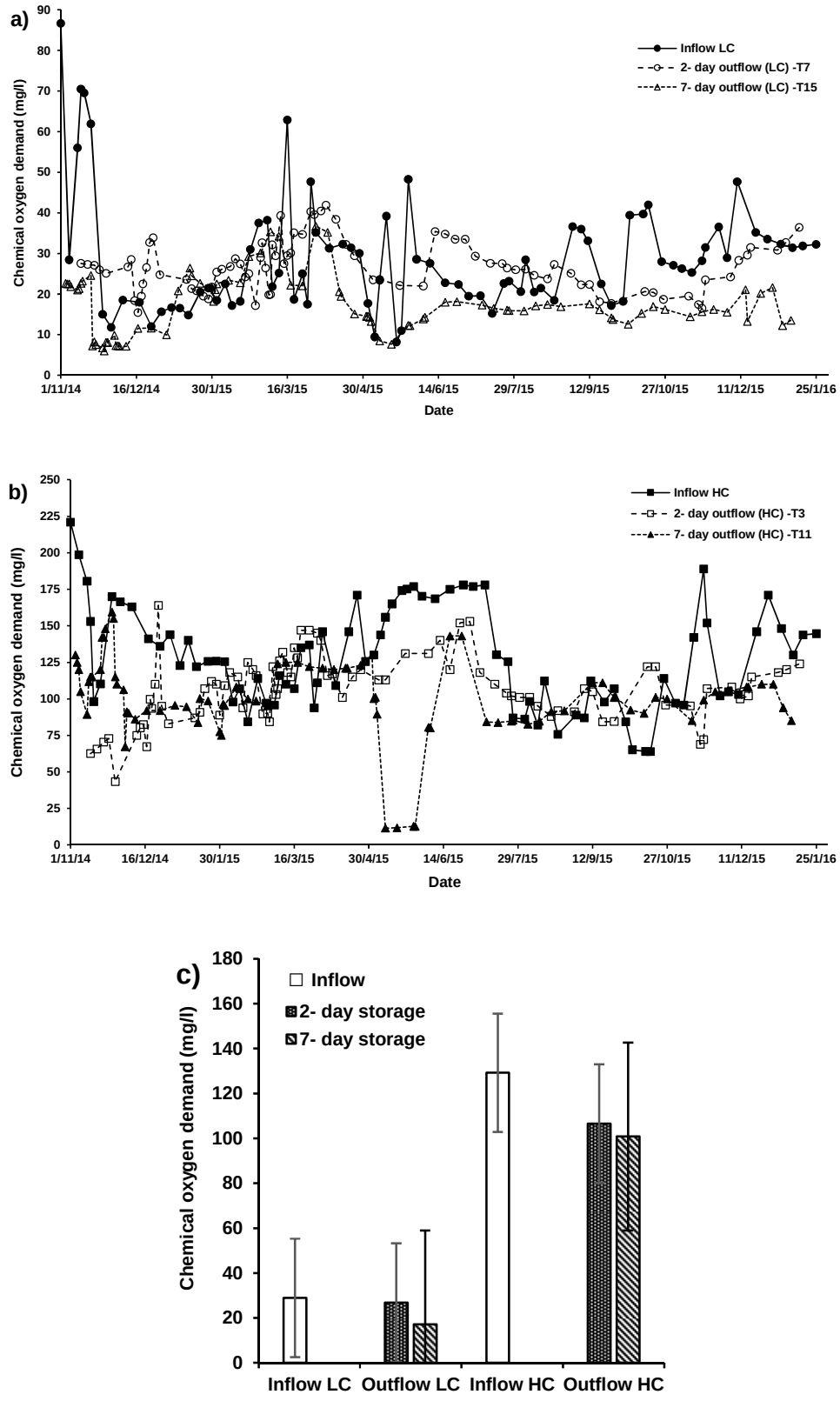


Figure 4.2. Effect of storage time on the variation of chemical oxygen demand for synthetic greywater in: a) low concentrations (LC), b) high concentrations (HC), and c) the average values for low and high concentrations.

For HC–SGW outflow, significantly ($p < 0.01$) negative correlations ($r = -0.315$, $p = 0.003$) were calculated between BOD_5 and COD for system T3, at 2–day contact time. However, after seven days of treatment, for the same design but for system T11, the correlations became significantly ($p < 0.01$) positive ($r = 0.398$, $p = 0.000$). Therefore, correlation of BOD_5 was significantly positively correlated with COD ($r = 0.306$, $p = 0.005$) for HC–SGW inflow.

In addition, the correlation analysis of wetlands treating LC–SGW showed no correlation between the outflow BOD_5 and COD for systems T7 (2–day storage time) and T15 (2–day storage time).

Moreover, the reduction in BOD_5 and COD concentrations of aquatic systems mainly depends upon the oxygen concentration in the outflow, as decomposition of pollutants occurs through oxidation reactions (Vymazal, 2010).

The BOD_5 to COD ratio of inflow SGW before utilisation in the experiment was 0.27 and 0.61 for HC–SGW and LC–SGW, respectively (Table 4.1).

The ratios of outflows were 0.14 for system T3 (2–day; HC–SGW), 0.21 for T7 (2–day; LC–SGW), 0.16 for T11 (7–day; HC–SGW) and 0.39 for T15 (7–day; LC–SGW). Similarly, low biodegradability of greywater was reported by Jefferson et al. (1999). However, after two hours of treatment, the BOD_5 to COD ratio of the greywater increased to 0.41, which is comparable with municipal wastewater, where the ratio is often between 0.4 and 0.8 (Marco et al., 1997). This demonstrates that the proportion of biodegradable contaminants in the treated water was higher than that in the raw greywater.

4.3.2. Nutrients

Table 4.3 shows the comparative statistical analysis between the inflow greywater characteristics and their corresponding ones for outflow. For HC–SGW, the NH_4–N concentrations show a stable behaviour with values around 0.4 mg/l without significant changes after two and seven days of treatment in systems T3 and T11, respectively. However, there were significant differences in outflow concentrations of NH_4–N in systems T7 and T15, which stored LC–SGW for 2– and 7–day duration times, respectively (Figure 4.3).

Increasing contact times insignificantly ($p > 0.05$) affected NH_4–N concentrations in the outflow of systems treating LC–SGW. A significant ($p < 0.05$) reduction in NH_4–N concentration was observed in the outflow of system T11 treating HC–SGW for seven days, compared to the system of 2–day contact time (T3), as shown in Table 4.4.

The outflow removals of NH_4–N for treatment systems containing LC–SGW were significantly ($p < 0.05$) higher than those removals of corresponding systems of HC–SGW for two days of contact time (T3 compared with T7) and seven days of contact time (T11 compared with T15).

The measured values for ammonia–nitrogen are close to the detection limit. Therefore, the transformation of ammonia to nitrite and subsequently to nitrate is not evident in this experiment (Essington, 2004). The extent of ammonia generation from the reduction of other nitrogenous forms such as NO_3–N and organic–N may have been substantial in accounting for the observed increases in some wetlands (Wallace et al., 2016).

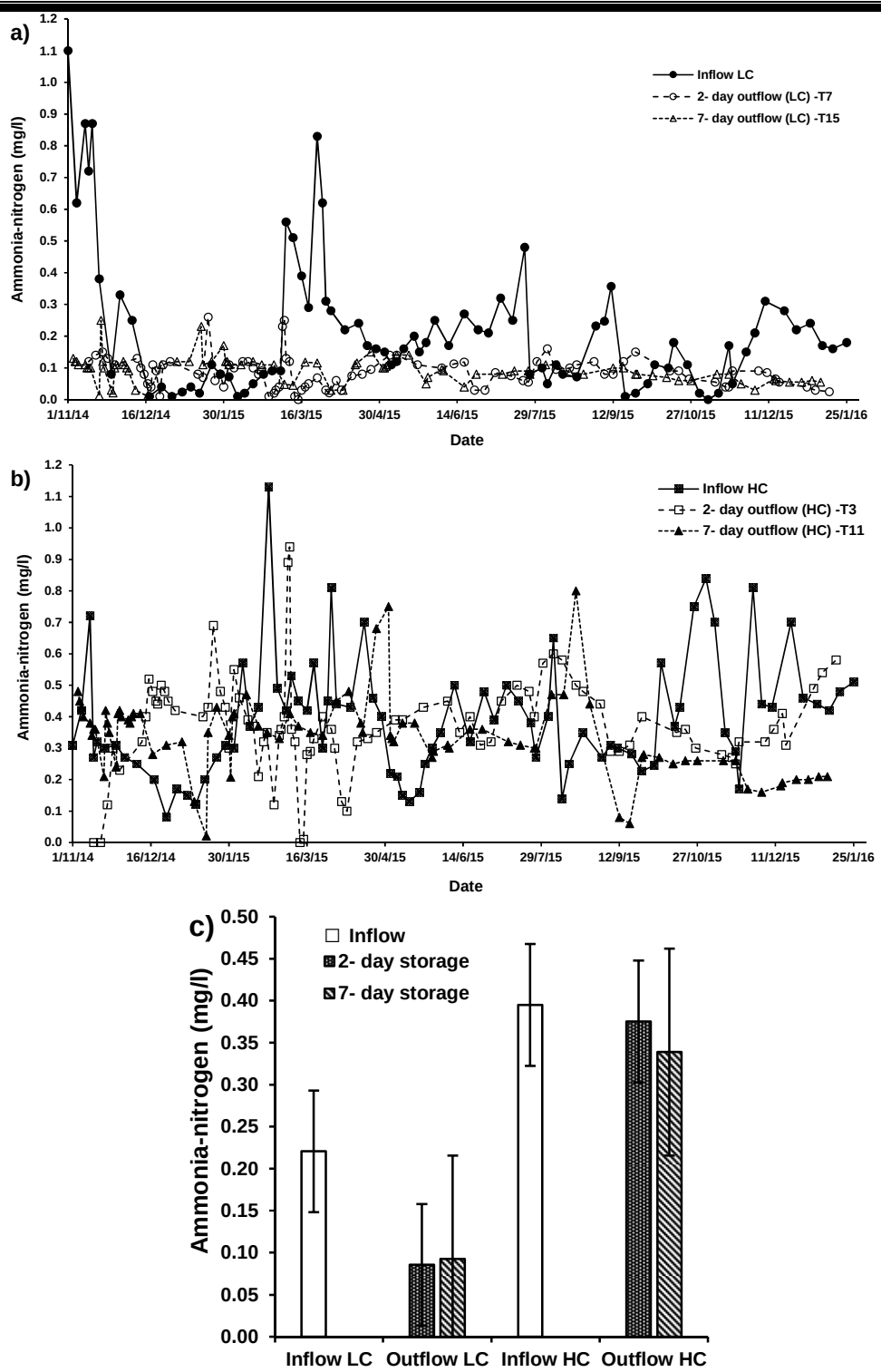


Figure 4.3. Effect of storage time on the variation of ammonia–nitrogen for synthetic greywater in: a) low concentrations (LC), b) high concentrations (HC), and c) the average values for low and high concentrations.

The correlation analysis indicated that the mean NH_4–N inflow concentration was significantly ($p < 0.01$) positively correlated ($r = 0.434$, $p = 0.000$) with COD of the LC–SGW inflow, while no correlation was observed between NH_4–N and BOD_5. While, the NH_4–N of outflow HC–SGW from system T11 (7–day contact time) was significantly ($p < 0.05$) positively correlated ($r = 0.219$, $p = 0.047$) with COD, no correlation with BOD_5 was indicated. Moreover, these correlation relationships were not found for T7 (2–day contact time).

For LC–SGW, the NH_4–N of system T7 (2–day contact time) was significantly ($p < 0.05$) negatively correlated ($r = -0.273$, $p = 0.012$) with COD. However, the NH_4–N of system T15 (7–day contact time) was significantly ($p < 0.05$) positively correlated ($r = 0.250$, $p = 0.023$) with BOD_5. While the NH_4–N outflow of C4 (containing tap water; 7–day contact time) was significantly ($p < 0.05$) negatively correlated (($r = -0.219$, $p = 0.047$) and ($r = -0.229$, $p = 0.037$)) with BOD_5 and COD, respectively, there was no correlation with the 2–day storage time of system C2.

No significant differences were indicated in comparison of the nitrate–nitrogen of inflow for either SGW with the outflows of all treatment systems (Table 4.3). A change was observed for the average values of nitrate–nitrogen after both storage times of HC–SGW. The values slightly increased from 8.9 mg/l to 9.4 mg/l after two days in system T3 and then decreased to reach 8.5 mg/l after seven days of storage time in system T11. The NO_3−N values of LC–SGW decreased slightly from 1.3 mg/l to 1.2 and to 1.0 mg/l after the two and seven days of storage time of systems T7 and T15, respectively, in this order (Tables 4.1, 4.2 and Figure 4.4).

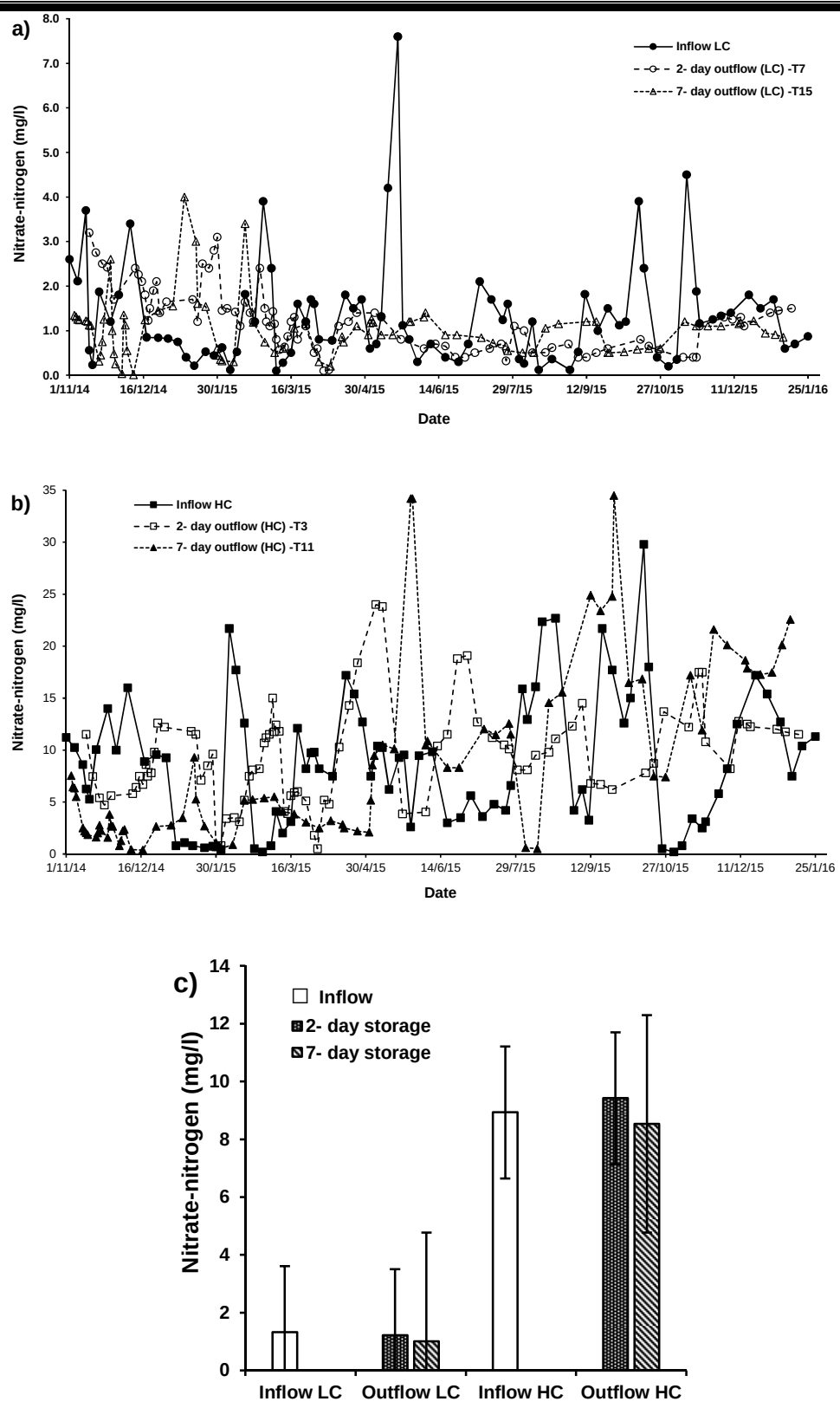


Figure 4.4. Effect of storage time on the variation of nitrate–nitrogen for synthetic greywater in: a) low concentrations (LC), b) high concentrations (HC), and c) the average values for low and high concentrations.

Increasing contact time had a significant effect in decreasing the NO_3–N concentrations in outflows from systems T11 (HC–SGW) and T15 (LC–SGW) compared to those values of the outflow of systems T3 (HC–SGW) and T7 (LC–SGW) at 2–day storage time. However, there were no significant differences in comparison of the NO_3–N removals between systems of HC–SGW (T3; 2–day and T11; 7–day) and of LC–SGW (T7; 2–day and T15; 7–day, respectively), as shown in Table 4.4.

The increase of nitrate–nitrogen can be explained by nitrification processes (Essington, 2004; Masi et al., 2010). However, an efficient removal of nitrogenous compounds, particularly NO_3−N, was because most greywaters have lack biodegradable organic matter, which can be a critical factor to utilise by denitrifying bacteria. This results commonly in low removal efficiencies regarding nitrogenous compounds in biological systems (Xiong et al., 2012; Cao et al., 2016).

In comparison of inflow and outflow ortho–phosphate–phosphorus (PO_4–P), significant decreases were observed in all treatment systems except for system T15 (7–day; LC–SGW), in which there was no significant change (Tables 4.1–4.3). Those decreased were from 59.1 mg/l for inflow to 46.2 mg/l (reduction of 21.8 %) for 2–day storage, and to 43.0 mg/l (reduction of 27.2 %) for 7–day storage, of HC–SGW. The decreases were from 8.4 mg/l for inflow to 7.0 mg/l for 2–day storage time, and slightly recovered to 8.5 mg/l for 7–day storage, of LC–SGW. Furthermore, the significant ($p < 0.05$) decreases in PO_4–P values with increasing storage time could be summarised by comparing the PO_4–P values after two days in systems T3 (HC–SGW) and T7 (LC–SGW) with those values after seven days of storage time in wetlands T11 (HC–SGW) and T15 (LC–SGW), respectively. However, only significant high removal of PO_4–P was found at 7–day storage time of LC–SGW in system T15 compared with system T11 (7–day; HC–SGW), as shown in Table 4.4 and Figure 4.5.

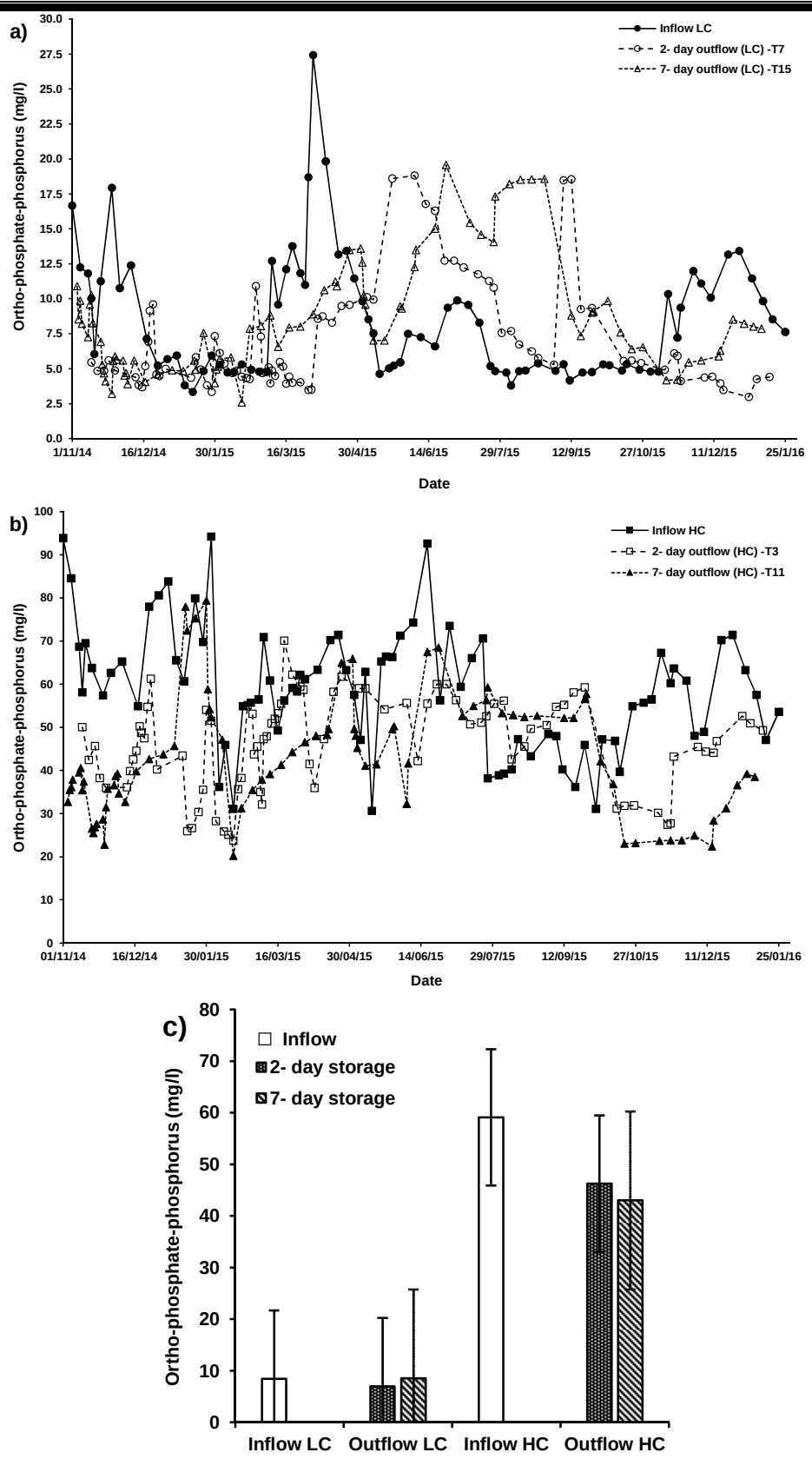


Figure 4.5. Effect of storage time on the variation of ortho–phosphate–phosphorus for synthetic greywater in: a) low concentrations (LC), b) high concentrations (HC), and c) the average values for low and high concentrations.

The HC–SGW and LC–SGW inflows were deficient and unbalanced in terms of nutrients and organic matter (Figure 4.6) due to the exclusion of urine and excreta (Abed and Scholz, 2016). An optimised nitrogen to phosphorus ratio can make a significant difference to the rates of oxygen uptake and COD removal for both real greywater and SGW (Al–Jayyousi, 2003). According to Jefferson et al. (2001), the deficiency of both macronutrients and trace nutrients in the greywater can limit the treatment efficiency of biological processes.

Table 4.5. Comparison of the nutrient balance ratio between the reported, inflow and outflow ratios of various set–up designs of experimental study.

	Inflow HC	***Outflow HC***		***Inflow LC***	***Outflow LC***		***Metcalf and Eddy Inc. (1991)****
	(mg/l)						
		T3	**T11**		**T7**	**T15**	(ratio)
P	59.1	46.2	43.0	8.4	7.0	8.5	1
N	9.3	9.8	8.8	1.5	1.29	1.1	20
COD	129.2	106.0	100.8	28.9	26.8	17.2	100

*Suggested COD : N : P ratio as 100 : 20 : 1 for wastewater.

The PO_4–P removals in systems can be explained by either sedimentation and/or microorganism activities. However, there are insufficient nutrients in LC–SGW for the survival of organisms, which are limited by PO_4–P removal (Jefferson et al., 2001). This scenario could be the case for system T15. Phosphorus is an essential nutrient for both plants and bacteria, although the portion of phosphorus, which is taken–up by plants is significantly lower than nitrogen (Brix, 1994). It is likely to be taken up by microbes developing in the outside systems (Friedler et al., 2006; Paulo et al., 2013).

However, given that microbes were not deliberately added to the greywater recipe, microbial biomass development is rather slow. Therefore, microbial activities accounted for only a small portion of the effects on phosphorus concentrations.

As reported, the settling of PO_4–P precipitates is a well–documented process in ponds, wetlands and similar systems (Ann et al., 1999; Peng et al., 2007) and was likely a contributing factor to the PO_4–P reduction throughout the systems. The uptake by algae and/or plants was also a likely factor in the PO_4–P decline. However, this is a seasonal process that would likely have reversed during plant die–off (Wallace et al., 2016).

4.3.3. Colour, Turbidity and Total Suspended Solids

In this study, outflow treated SGW was agitated before sampling to simulate the washing–out of solids, possibility to slough away biofilm attached to the walls of the treatment vessel (Zhang et al., 2015), as well as to encourage the solids to dissolve and/or re–suspend into the water column (Stewart et al., 2008; Van de Moortel et al., 2010; Tanner and Headley, 2011).

However, data show a reduction in colour, turbidity and total suspended solids for the outflow of 2–day storage experiments by 23.3%, 11.8% and 24.8%, respectively. The percentages concerning the outflow for the 7–day storage experiments were 28.9%, 27.9% and 37.3%, respectively. The number of colloids and particles is likely to reduce over time as physical (e.g., coagulation and flocculation) processes reduce turbidity and suspended solids (Nolde, 1999).

However, biochemical processes such as biodegradation will lead to an increase in microorganisms and debris contributing to an increase in turbidity and fine material (Christova–Boal et al., 1996; Dixon et al., 1999; Günther, 2000; Eriksson et al., 2002; Gross et al., 2006; Gross et al., 2007; F. Li et al., 2009).

However, a statistical analysis showed no significant ($p > 0.05$) changes in colour, pH, turbidity and total suspended solids, when both synthetic greywaters were stored for two or seven days. This confirms previous findings (Diaper et al., 2008; Hourlier et al., 2010) showing that suspended solids and insoluble particle concentrations of chemical greywaters are highly stable, possibly, because they originate from inert materials.

For HC–SGW, treatment system T11 showed significant ($p < 0.05$) drops in turbidity at seven days of contact compared to the system of 2–day contact time. In treatment of LC–SGW, outflow means of TSS and turbidity decreased significantly in outflows of systems T7 and T15, compared with those values of inflow (Table 4.3).

Increasing treatment time had a significant ($p < 0.05$) effect, decreasing TSS concentrations in the outflows of system T11 treating HC–SGW compared to those concentrations in system T3 (2–day contact time).

However, no significant ($p > 005$) effect of treatment time in terms of turbidity was calculated for either system. The TSS and turbidity values decreased significantly ($p < 0.05$) with increasing treatment time in the outflows of LC–SGW treated by system T15 compared to system T7 operating at 2–day contact time (Table 4.4).

A comparison of the outflows of systems treating HC–SGW (T3 and T11), in terms of TSS and turbidity removals, were significantly ($p < 0.05$) lower than those removals for corresponding systems treating LC–SGW (T7 and T15,) respectively at both contact times (Table 4.5).

4.3.4. Minerals and Trace Elements in Inflow/Outflow

The chemical analysis for trace element concentrations of inflow SGW are shown in Table 4.5.

Table 4.6. The chemical analysis of trace element concentrations (mg/l) of inflow low (LC) and high (HC) pollutant content synthetic greywater (SGW).

Element	Inflow (low concentration)					Inflow (high concentration)				
	n	Mean	SD	Min	Max	n	Mean	SD	Min	Max
Aluminium (Al)	45	0.52	0.528	0.09	1.56	45	2.13	0.869	0.76	4.77
Boron (B)	33	0.14	0.067	0.10	0.36	33	0.57	0.068	0.49	0.72
Calcium (Ca)	55	10.54	0.853	8.51	11.81	45	36.08	8.750	22.38	50.72
Cadmium (Cd)	42	0.09	0.056	0.04	0.23	39	7.36	2.981	3.67	11.58
Chromium (Cr)	54	0.04	0.063	0.01	0.23	54	3.20	0.918	1.67	5.70
Copper (Cu)	63	0.16	0.058	0.10	0.30	63	1.44	0.435	0.95	2.03
Iron (Fe)	48	0.21	0.102	0.11	0.43	48	6.41	2.476	2.48	10.37
Potassium (K)	12	4.04	0.448	3.41	4.65	12	60.16	1.684	58.31	62.53
Magnesium (Mg)	48	1.45	0.191	1.19	1.78	48	17.16	2.119	12.68	20.33
Manganese (Mn)	63	0.17	0.084	0.07	0.26	63	0.98	0.257	0.64	1.29
Sodium (Na)	12	14.32	1.662	12.01	16.00	12	62.68	14.538	45.50	76.92
Nickel (Ni)	51	0.04	0.065	0.01	0.18	51	0.05	0.065	0.01	0.20
Zinc (Zn)	39	0.21	0.159	0.01	0.39	39	4.25	1.500	2.22	6.34

Note: n, number of tested samples; SD, standard deviation; Min, minimum; Max, maximum.

Furthermore, the outflow SGW were elementally analysed using ICP_OES and the results are summarised in Table 4.6.

The statistical comparisons between the inflow and the outflow SGW, in terms of trace element concentrations, showed no significant changes in concentrations of Fe, Mg and Na in all treatment systems. Furthermore, the same comparison strategy revealed there were also no significant differences in concentrations of B, Cd, Cu and Zn in outflow of HC–SGW systems (T3 and T11), and Al, Cr, K, and Mn in outflow of LC–SGW systems (T7 and T15) for both times of treatments compared to those values in the inflow SGW. Also, concentrations of Al and Cr (T3 and T7; 2–day contact time) and Ca, Cd and Zn (T11 and T15; 7–day contact time) were not significantly affected in treatment systems treating both types of SGW (Table 4.7 and Figure 4.7). Concentrations of B and

Cu (T7 and T15; LC–SGW) and K (T3 and T11; HC–SGW) decreased significantly for both treatment times.

Considerable decreases in concentrations of Cd, Cr, Ni and Zn were indicated in the outflow of system T7 treating LC–SGW for two days with significant increasing of Ca and Fe concentrations in that system, compared with their contents in the inflow. In addition, only a significant increase in Ni content was indicated, after treating LC–SGW for seven days in system T15.

For HC–SGW, significant increases were noted in outflow concentrations of Ca (T3; 2–day), Al and Ni (T11; 7–day), and Cr and Mn (both times) compared to those contents in the inflow.

Table 4.7. The chemical analysis of trace element concentrations (mg/l) of outflow low (LC) and high (HC) pollutant content synthetic greywater (SGW) treated for 2– and 7–day contact times.

Element	2–day outflow (HC)–T3						2–day outflow (LC)–T7					
	n	Mean	SD	Min	Max	Rem (%)	n	Mean	SD	Min	Max	Rem (%)
Aluminium	39	2.41	1.016	0.74	4.25	-13.15	39	0.34	0.180	0.11	0.72	34.62
Boron	35	0.54	0.060	0.42	0.66	5.26	35	0.11	0.009	0.08	0.13	21.43
Calcium	37	43.02	2.411	35.94	46.28	-19.24	46	11.25	0.773	9.86	12.70	-6.74
Cadmium	42	7.69	1.064	4.95	8.98	-4.48	42	0.05	0.031	0.00	0.11	44.44
Chromium	58	3.76	1.203	1.34	4.98	-17.5	58	0.04	0.049	0.00	0.12	0.00
Copper	63	1.45	0.113	1.28	1.70	-0.69	63	0.06	0.049	0.02	0.15	62.50
Iron	51	6.35	2.423	1.56	9.29	0.94	51	0.21	0.157	0.09	0.45	0.00
Potassium	14	55.68	4.486	49.48	60.69	7.45	14	3.87	0.364	3.35	4.50	4.21
Magnesium	48	17.76	1.392	13.92	19.55	-0.20	48	1.35	0.133	0.99	1.58	6.90
Manganese	63	1.19	0.063	1.06	1.29	-21.4	63	0.08	0.056	0.00	0.18	52.94
Sodium	14	58.19	10.620	42.35	68.22	7.16	14	13.82	1.175	12.14	15.57	3.49
Nickel	53	0.03	0.018	0.00	0.06	40.00	53	0.01	0.007	0.00	0.04	75.00
Zinc	44	4.30	0.524	3.12	5.25	-1.42	42	0.09	0.083	0.00	0.23	57.14
	7–day outflow (HC)–T11						7–day outflow (LC)–T15					
Aluminium	54	2.98	1.218	1.61	6.14	-39.91	44	0.36	0.189	0.09	0.75	30.77
Boron	26	0.54	0.160	0.34	0.77	5.26	24	0.12	0.064	0.08	0.26	14.29
Calcium	52	37.39	4.030	30.58	45.66	-3.63	52	10.74	0.739	9.44	12.12	-1.90
Cadmium	36	6.40	1.984	3.86	9.72	13.59	32	0.09	0.083	0.00	0.21	0.00
Chromium	46	4.76	1.215	2.83	6.68	-48.75	42	0.07	0.074	0.00	0.21	-75.00
Copper	54	1.30	0.301	0.80	1.76	9.72	48	0.10	0.091	0.00	0.26	37.50
Iron	42	7.02	1.801	3.58	9.36	-9.52	38	0.20	0.100	0.07	0.30	4.76
Potassium	8	45.77	5.160	39.87	51.00	23.92	8	3.62	0.438	3.07	4.22	10.40
Magnesium	54	16.24	1.971	11.76	18.35	5.36	46	1.38	0.161	1.03	1.64	4.83
Manganese	54	1.01	0.223	0.75	1.38	-3.06	48	0.06	0.074	0.00	0.21	64.71
Sodium	8	55.22	11.852	41.86	67.68	11.90	8	13.15	1.199	11.83	14.36	8.17
Nickel	50	0.09	0.081	0.00	0.20	-80.00	44	0.05	0.080	0.00	0.18	-25.00
Zinc	32	3.90	0.972	1.90	5.10	8.02	28	0.13	0.068	0.01	0.25	38.10

Note: n, number of tested samples; SD, standard deviation; Min, minimum; Max, maximum; Rem, removal.

Table 4.8. Significant values of the statistical analysis concerning the comparison between the inflow and outflow greywater in terms of trace element concentrations (mg/l).

	2–day contact time						7–day contact time					
	Inflow & T3 (HC–SGW)			*Inflow & T7 (LC–SGW)*			*Inflow & T11 (HC–SGW)*			*Inflow & T15 (LC–SGW)*		
Element	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
Aluminium	0.009	M–W	0.171	<0.001	M–W	0.192	0.156	T–test	<0.001	<0.001	M–W	0.321
Boron	0.002	M–W	0.171	<0.001	M–W	0.001	0.016	M–W	0.593	<0.001	M–W	<0.001
Calcium	0.001	M–W	0.001	0.058	T–test	<0.001	0.003	M–W	0.068	0.003	M–W	0.295
Cadmium	0.001	M–W	0.159	<0.001	M–W	<0.001	<0.001	M–W	0.340	<0.001	M–W	0.290
Chromium	0.004	M–W	0.003	<0.001	M–W	0.014	<0.001	M–W	<0.001	<0.001	M–W	0.360
Copper	<0.001	M–W	0.378	<0.001	M–W	<0.001	<0.001	M–W	0.177	<0.001	M–W	<0.001
Iron	0.002	M–W	0.867	<0.001	M–W	0.001	0.003	M–W	0.193	<0.001	M–W	0.444
Potassium	0.004	M–W	0.013	0.125	T–test	0.309	0.002	M–W	<0.001	0.334	T–test	0.054
Magnesium	0.007	M–W	0.120	<0.001	M–W	0.776	0.024	M–W	0.277	<0.001	M–W	0.531
Manganese	<0.001	M–W	0.006	<0.001	M–W	0.250	<0.001	M–W	0.023	<0.001	M–W	0.875
Sodium	0.007	M–W	0.322	0.010	M–W	0.403	0.005	M–W	0.181	0.012	M–W	0.181
Nickel	<0.001	M–W	0.169	<0.001	M–W	<0.001	<0.001	M–W	0.016	<0.001	M–W	<0.001
Zinc	0.005	M–W	0.500	<0.001	M–W	<0.001	0.001	M–W	0.503	<0.001	M–W	0.328

Note: HC, high concentration; LC, low concentration; SGW, synthetic greywater p value, significant difference if $p < 0.05$ and not significant if $p > 0.05$; M–W, Mann–Whitney U–test.

Table 4.9. Significant values of the statistical analysis for outflow greywater after 2– and 7–day storage time to investigate the effects of increasing contact time and pollutant loads on changing the trace element concentrations (mg/l).

	Effects of increasing contact time						Effects of pollutant loads					
	HC–SGW			*LC–SGW*			*2–day contact time*			*7–day contact time*		
	T3 & T11			T7 & T15			T3 & T7			T11 & T15		
Element	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
Aluminium	0.329	T–test	0.020	<0.001	M–W	0.906	<0.001	M–W	0.213	<0.001	M–W	0.064
Boron	0.003	M–W	0.815	<0.001	M–W	0.001	<0.001	M–W	0.035	<0.001	M–W	0.038
Calcium	<0.001	M–W	<0.001	0.001	M–W	0.002	<0.001	M–W	<0.001	<0.001	M–W	0.265
Cadmium	0.002	M–W	0.006	<0.001	M–W	0.243	0.003	M–W	<0.001	<0.001	M–W	0.201
Chromium	0.002	M–W	<0.001	<0.001	M–W	0.557	0.004	M–W	0.039	<0.001	M–W	0.274
Copper	<0.001	M–W	<0.001	<0.001	M–W	0.630	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Iron	<0.001	M–W	0.089	<0.001	M–W	0.285	<0.001	M–W	0.221	<0.001	M–W	0.003
Potassium	0.016	M–W	<0.001	0.891	T–test	0.161	0.002	M–W	0.729	0.103	T–test	0.001
Magnesium	<0.001	M–W	<0.001	<0.001	M–W	0.314	0.016	M–W	0.005	0.171	T–test	0.866
Manganese	<0.001	M–W	0.001	<0.001	M–W	0.024	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Sodium	0.009	M–W	0.267	0.044	M–W	0.441	0.026	M–W	0.488	0.009	M–W	0.009
Nickel	<0.001	M–W	0.001	<0.001	M–W	0.287	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Zinc	<0.001	M–W	0.130	<0.001	M–W	0.057	<0.001	M–W	<0.001	<0.001	M–W	0.398

Note: HC, high concentration; LC, low concentration; SGW, synthetic greywater; p value, significantly different if $p < 0.05$ and not significantly different if $p > 0.05$; M–W, Mann–Whitney U–test.

To study the effect of increasing contact time on the trace element concentrations, outflows from different systems at 7–day contact time were statistically compared to outflows of 2–day contact time. There were no significant changes in concentrations of Al, B, Cd, Cr, Cu, Fe, K, Mg, Na, Ni and Zn in treatment systems of LC–SGW, and concentrations of B, Fe, Na and Zn in treatment system of HC–SGW, with increasing contact time. However, increasing contact time had a significant effect in decreasing the outflow concentrations of Ca and Mn (LC–SGW) and Ca, Cd, Cu, K, Mg and Mn (HC–SGW) after 7–day treatment time compared with systems of 2–day treatment time. With increasing contact time, only significant increases in concentrations of Al, Cr and Ni were indicated in the outflow of system T15 (HC–SGW; 7–day) (Table 4.8 and Figure 4.7).

The possible mechanisms of pollutant removals in systems such as these are either sedimentation and/or biodegradation due to microorganism activities. However, the SGW was agitated before sampling to simulate the washing–out of solids, resuspension of solids, and biofilm slough–out from the treatment vessel wall. Therefore, the development of microorganisms could be the dominating mechanism.

The significant changes in most trace element concentrations were observed to be higher in systems treating LC–SGW than those of HC–SGW. The reason for this could be due to the high solid contents, high values of pH and other pollutant strengths which presented in HC–SGW, as well as the boundary conditions of those systems which directly subjected to the environment.

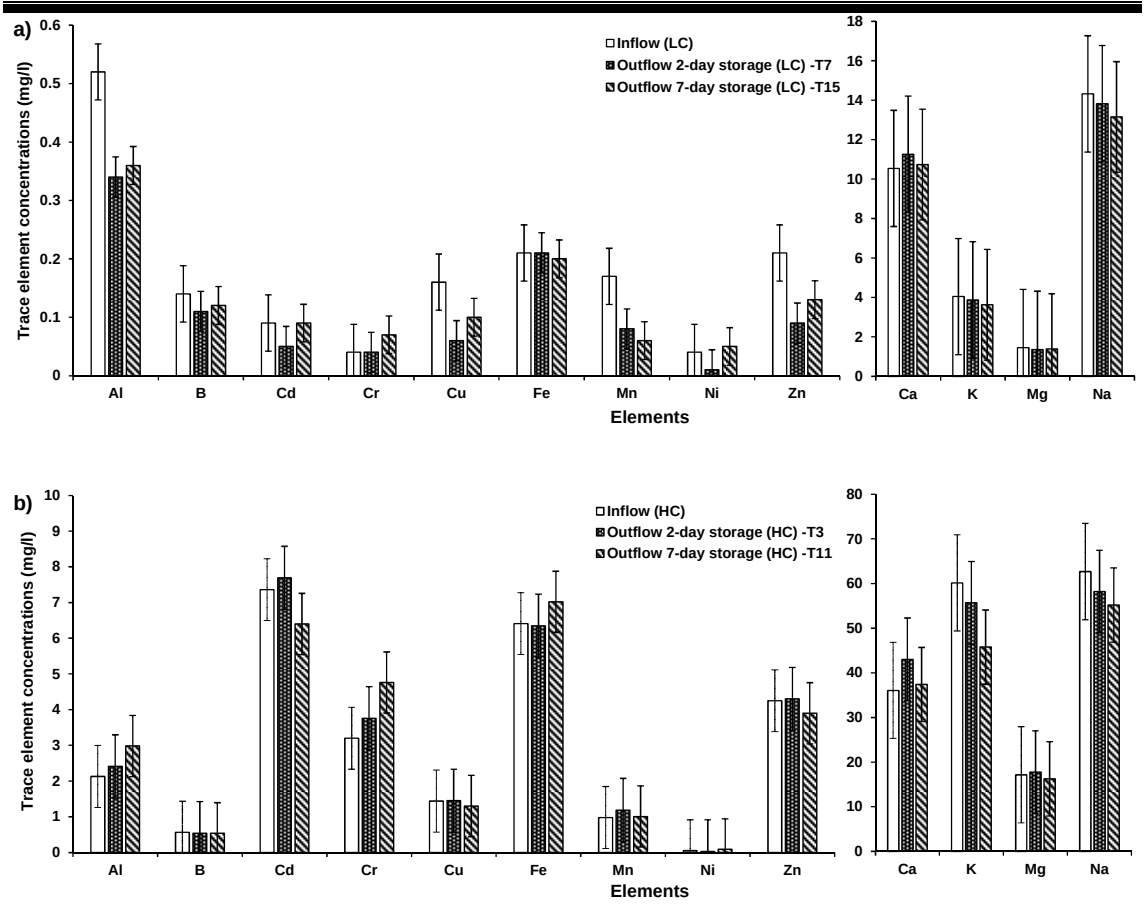


Figure 4.6. Effect of storage time on the variation in the average concentrations of trace elements in both types of synthetic greywater: *a)* low concentrations (LC), *b)* high concentrations (HC).

Chapter Five

Phytoremediation of Synthetic Greywater in Mesocosm Scale Floating Treatment Wetlands (FTWs)

Abed, S. N., Almuktar, S.A.A.A.N., Scholz, M., 2017. Remediation of synthetic greywater in mesocosm–scale floating treatment wetlands. Ecological Engineering, 102, 303–319.

5.1. Overview

In chapter five, mesocosm systems contained only synthetic greywater and those treated the synthetic greywaters with only floating *P. australis* (T1, T3, T5, T7, T0, T11, T13 and T16) were considered as a simulation of natural floating reedbed or floating treatment wetlands and mentioned as (F1, F2, F3, F4, F5, F6, F7 and F8, respectively). Furthermore, control wetlands which receiving clean tap water with (C1 and C3) and without (C2 and C4) floating *P. australis* were also considered, as shown in Table 5.1.

Table 5.1. Operational parameters in experimental set–up designs for different floating treatment wetlands.

Wetland[a]	Original label	Contact time		Water type			Vegetation	
		Two days	*Seven days*	*High concentration*	*Low concentration*	*TW[b]*	*With*	*Without*
F1	T1	✓		✓			✓	
F2	T3	✓		✓				✓
F3	T5	✓			✓		✓	
F4	T7	✓			✓			✓
F5	T9		✓	✓			✓	
F6	T11		✓	✓				✓
F7	T13		✓		✓		✓	
F8	T15		✓		✓			✓
C1	C1	✓				✓	✓	
C2	C2	✓				✓		✓
C3	C3		✓			✓	✓	
C4	C4		✓			✓		✓

[a] Wetlands from F1 to F8 have four replicates, while C1 to C4 have two replicates.
[b] Tap water.

The inflow water quality was compared with the quality of outflows from each floating treatment wetland to study their performance. The effect of experimental variables such as presence of *P. australis*, contact time and concentrations of pollutant were investigated by comparison of outflows from treatment systems of floating *P. australis*, F1 (T1), F3 (T5), F5 (T9) and F7 (T13) with each other, from one side, and with outflows of corresponding treatment system which contained only greywater, F2 (T3), F4 (T7), F6 (T11) and F8 (T15), respectively, which discussed in chapter four. The focus on removal processes (other than sedimentation) such as the role of macrophytes in phytoremediation was achieved by agitation of the treated outflow before sampling. To assess the impact of interactions between the experimental operational variables on removal performances of FTW, different approaches of statistical analysis were applied.

To cover objective 2 of this study, section 1.5, different statistical analysis techniques were subjected on the outflow water parameters in terms of significant differences and correlation relationship. Oxygen demands were studied in terms of chemical and biochemical (COD and BOD_5, respectively) and the correlation between them into inflow and outflow. Biodegradation was discussed and identified as a ratio between them. Nutrient concentrations and nutrient balance, suspended solids and other parameter were investigated as well. *P. australis* growth rate in each floating treatment wetland was monitored and discussed at the end on this chapter.

5.2. Inflow Greywater Quality

Table 5.2 shows the overall inflow water quality of both LC–SGW and HC–SGW for different loads of pollutants, which discussed in chapter four. Various outflow parameters were compared to assess potentially significant differences to highlight the

effect of experimental operational parameters such as the presence of floating *P. australis*, increase treatment contact time and the load pollutant contents in greywater.

Table 5.2. Physiochemical characteristics of inflow synthetic greywater (SGW).

Parameter	*Inflow (Low concentration)*						*Inflow (High concentration)*				
	Unit	n	Mean	SD	Min	Max	n	Mean	SD	Min	Max
pH	–	81	6.9	0.48	5.3	7.9	81	8.4	1.61	5.4	11.5
Redox potential	mV	81	34.1	21.23	-18.1	111.2	81	-36.6	74.22	-182.1	97.9
Turbidity	NTU	81	22.9	7.14	9.8	41.6	81	188.9	47.22	18.3	308.0
Total suspended solids	mg/l	81	39.9	15.94	10.0	87.0	81	317.0	58.35	173.0	473.0
Electrical conductivity	µS/cm	81	164.6	63.24	98.7	452.0	81	988.5	196.09	612.0	1677.0
Dissolved oxygen	mg/l	81	10.4	1.24	7.7	12.3	81	10.5	1.39	6.9	12.6
Colour	Pa/Co	81	214.5	64.07	26.0	340.0	81	1587.8	379.89	787.0	2499.0
Temperature	°C	81	17.7	4.58	6.7	27.0	81	16.9	5.40	6.5	27.8
Biochemical oxygen demand	mg/1	81	17.6	8.00	2.0	40.0	81	34.7	12.99	10.0	60.0
Chemical oxygen demand	mg/1	81	28.9	14.47	8.2	86.7	81	129.2	34.68	63.9	221.0
Ammonia–nitrogen	mg/1	81	0.2	0.22	0.0	1.1	81	0.4	0.19	0.1	1.1
Nitrate–nitrogen	mg/1	81	1.3	1.21	0.1	7.6	81	8.9	6.38	0.2	29.8
Ortho–phosphate–phosphorus	mg/1	81	8.4	4.36	3.3	27.4	81	59.1	14.16	30.6	94.2

Note: n, number of tested samples; SD, standard deviation; Min, minimum; Max, maximum; and NTU, nephelometric turbidity unit.

5.3. Outflow Greywater Quality

Characteristics of the outflow greywater quality are shown in Tables 5.3 and 5.4. The outflow quality of the control floating wetlands containing tap water with retention times of two and seven days is summarised in Table 5.5.

Due to the difficulties in operation of FTWs (highly complex open systems), they are subject to ambient environmental conditions and a number of interacting processes (Wallace et al., 2016). Treated SGW outflows were statistically compared with corresponding values of the inflow. To assess the role of floating macrophytes, values of greywater parameters of wetlands treating HC–SGW and LC–SGW with floating *P. australis* for 2–day contact times (i.e. F1 and F3, respectively) were compared with corresponding values of outflows from wetlands F2 and F4, where the design set–up was the same but plants were absent.

Table 5.3. Wetland (F) outflow parameters for greywater with high concentration (HC) and low concentration (LC) after 2–day contact time.

Parameter	Unit	n	Mean	SD	Min	Max	Rem. (%)
2–day outflow (HC)–F1							
pH	–	85	7.4	1.09	5.7	9.5	na
Redox potential	mV	85	8.1	52.68	-89.5	87.2	na
Turbidity	NTU	85	175.9	59.61	39.8	319.0	6.9
Total suspended solids	mg/l	85	302.9	75.19	79.0	455.0	4.4
Electrical conductivity	µS/cm	85	987.4	107.25	713.0	1225.0	na
Dissolved oxygen	mg/l	85	9.0	1.03	5.5	11.9	14.3
Colour	Pa/Co	85	1525.6	411.54	455.0	2361.0	3.9
Temperature	°C	85	17.1	4.92	5.8	24.6	na
Biochemical oxygen demand	mg/l	85	17.7	6.40	0.0	30.0	49.0
Chemical oxygen demand	mg/l	85	96.3	32.01	33.8	172.0	25.5
Ammonia–nitrogen	mg/l	85	0.4	0.21	0.0	1.2	0.0
Nitrate–nitrogen	mg/l	85	14.1	6.40	0.2	32.0	-58.4
Ortho–phosphate–phosphorus	mg/l	85	52.0	14.87	15.8	95.5	12.0
2–day outflow (HC)–F2							
pH	–	85	7.8	1.37	5.6	9.8	na
Redox potential	mV	85	-3.0	62.95	-107.6	88.6	na
Turbidity	NTU	85	192.1	50.87	102.0	341.0	-1.7
Total suspended solids	mg/l	85	321.8	56.68	165.0	447.0	-1.5
Electrical conductivity	µS/cm	85	965.2	106.68	627.0	1208.0	na
Dissolved oxygen	mg/l	85	10.2	0.73	8.7	12.1	2.9
Colour	Pa/Co	85	1527.6	326.28	677.0	2311.0	3.8
Temperature	°C	85	17.1	4.75	6.1	23.6	na
Biochemical oxygen demand	mg/l	85	14.7	7.78	0.0	40.0	57.6
Chemical oxygen demand	mg/l	85	106.6	22.68	43.3	164.0	17.5
Ammonia–nitrogen	mg/l	85	0.4	0.16	0.0	0.9	0.0
Nitrate–nitrogen	mg/l	85	9.4	4.67	0.5	24.0	-5.6
Ortho–phosphate–phosphorus	mg/l	85	46.2	10.74	23.7	70.1	21.8
2–day outflow (LC)–F3							
pH	–	85	7.0	0.71	6.1	10.2	na
Redox potential	mV	85	27.5	32.18	-120.8	72.4	na
Turbidity	NTU	85	28.2	37.09	3.4	262.0	-23.1
Total suspended solids	mg/l	85	41.7	43.57	8.0	316.0	-4.5
Electrical conductivity	µS/cm	85	145.9	30.41	15.5	325.0	na
Dissolved oxygen	mg/l	85	9.3	1.08	6.9	12.5	10.6
Colour	Pa/Co	85	183.7	74.89	73.0	476.0	14.4
Temperature	°C	85	17.0	4.84	6.0	23.0	na
Biochemical oxygen demand	mg/l	85	9.9	5.49	0.0	30.0	43.8
Chemical oxygen demand	mg/l	85	32.4	14.55	10.6	89.6	-13.8
Ammonia–nitrogen	mg/l	85	0.1	0.07	0.0	0.3	50.0
Nitrate–nitrogen	mg/l	85	1.7	1.13	0.0	5.8	-30.8
Ortho–phosphate–phosphorus	mg/l	85	7.6	3.90	3.2	120	9.5
2–day outflow (LC)–F4							
pH	–	85	7.5	0.70	6.3	10.1	na
Redox potential	mV	85	4.2	30.40	-116.1	51.0	na
Turbidity	NTU	85	20.2	14.20	2.9	129.0	11.8
Total suspended solids	mg/l	85	30.0	12.12	11.0	76.0	24.8
Electrical conductivity	µS/cm	85	138.5	23.26	79.0	215.0	na
Dissolved oxygen	mg/l	85	10.5	0.82	8.2	12.6	-1.0
Colour	Pa/Co	85	164.5	40.93	34.0	265.0	23.3
Temperature	°C	85	16.0	4.59	5.3	21.8	na
Biochemical oxygen demand	mg/l	85	5.6	3.60	0.0	20.0	68.2
Chemical oxygen demand	mg/l	85	26.8	6.18	15.4	41.9	7.3
Ammonia–nitrogen	mg/l	85	0.09	0.05	0.0	0.3	55.0
Nitrate–nitrogen	mg/l	85	1.2	0.71	0.1	3.2	7.7
Ortho–phosphate–phosphorus	mg/l	85	7.0	3.89	3.0	18.8	16.7

Note: n, number of tested samples; SD, standard deviation; Min, minimum; Max, maximum; Rem., removal; and NTU, nephelometric turbidity unit.

Table 5.4. Wetland (F) outflow parameters for greywater with high concentration (HC) and low concentration (LC) after 7–day contact time.

Parameter	Unit	n	Mean	SD	Min	Max	Rem. (%)
7–day outflow (HC)–F5							
pH	–	83	7.3	0.82	5.9	8.8	na
Redox potential	mV	83	12.2	40.30	-57.6	77.5	na
Turbidity	NTU	83	154.8	86.08	9.8	430.0	18.1
Total suspended solids	mg/l	83	267.8	110.05	26.0	458.0	15.5
Electrical conductivity	µS/cm	83	1137.4	471.09	475.0	3010.0	na
Dissolved oxygen	mg/l	83	8.8	0.89	6.5	11.0	16.2
Colour	Pa/Co	83	1448.1	647.98	106.0	2941.0	8.8
Temperature	°C	83	16.8	4.03	7.2	22.5	na
Biochemical oxygen demand	mg/l	83	23.1	9.35	8.0	40.0	33.4
Chemical oxygen demand	mg/l	83	94.0	31.13	16.2	161.5	27.2
Ammonia–nitrogen	mg/l	83	0.5	0.23	0.2	1.2	-25.0
Nitrate–nitrogen	mg/l	83	10.7	7.92	0.9	37.9	-20.2
Ortho–phosphate–phosphorus	mg/l	83	48.0	13.76	23.1	76.3	18.9
7–day outflow (HC)–F6							
pH	–	83	7.7	1.21	5.9	9.9	na
Redox potential	mV	83	-4.4	59.67	-108.3	78.1	na
Turbidity	NTU	83	185.7	49.24	65.1	281.0	1.7
Total suspended solids	mg/l	83	302.6	61.44	147.0	434.0	4.5
Electrical conductivity	µS/cm	83	1003.0	306.88	492.0	2460.0	na
Dissolved oxygen	mg/l	83	10.5	0.91	7.9	12.0	0.0
Colour	Pa/Co	83	1644.8	489.96	718.0	2889.0	-3.6
Temperature	°C	83	16.6	3.87	7.6	22.1	na
Biochemical oxygen demand	mg/l	83	16.6	7.07	4.0	38.0	52.2
Chemical oxygen demand	mg/l	83	100.8	27.65	11.6	159.5	22.0
Ammonia–nitrogen	mg/l	83	0.3	0.13	0.0	0.8	25.0
Nitrate–nitrogen	mg/l	83	8.5	8.42	0.4	34.5	4.5
Ortho–phosphate–phosphorus	mg/l	83	43.0	13.78	20.25	79.4	27.2
7–day outflow (LC)–F7							
pH	–	83	6.9	0.61	6.2	9.0	na
Redox potential	mV	83	31.0	28.12	-75.5	68.9	na
Turbidity	NTU	83	18.9	11.05	2.8	52.1	17.5
Total suspended solids	mg/l	83	27.7	16.48	2.8	72.0	30.6
Electrical conductivity	µS/cm	83	161.4	42.91	103.7	324.0	na
Dissolved oxygen	mg/l	83	9.3	1.24	4.9	12.0	10.6
Colour	Pa/Co	83	159.1	56.83	48.0	391.0	25.8
Temperature	°C	83	15.9	4.18	7.3	22.4	na
Biochemical oxygen demand	mg/l	83	13.4	5.63	2.0	30.0	23.9
Chemical oxygen demand	mg/l	83	31.3	11.95	7.9	77.5	-8.3
Ammonia–nitrogen	mg/l	83	0.1	0.07	0.0	0.5	50.0
Nitrate–nitrogen	mg/l	83	1.3	0.77	0.0	4.8	0.0
Ortho–phosphate–phosphorus	mg/l	83	11.9	6.36	3.3	32.0	-41.7
7–day outflow (LC)–F8							
pH	–	83	7.5	0.72	6.4	9.3	na
Redox potential	mV	83	1.8	33.00	-87.9	53.2	na
Turbidity	NTU	83	16.5	7.27	5.7	34.1	27.9
Total suspended solids	mg/l	83	25.0	10.96	7.0	56.0	37.3
Electrical conductivity	µS/cm	83	144.0	32.28	97.7	263.0	na
Dissolved oxygen	mg/l	83	11.0	1.11	8.1	14.3	-5.8
Colour	Pa/Co	83	152.6	41.05	51.0	258.0	28.9
Temperature	°C	83	15.3	4.23	6.7	22.2	na
Biochemical oxygen demand	mg/l	83	6.7	4.85	0.0	22.0	61.9
Chemical oxygen demand	mg/l	83	17.2	6.95	6.0	36.7	40.5
Ammonia–nitrogen	mg/l	83	0.1	0.04	0.0	0.3	50.0
Nitrate–nitrogen	mg/l	83	1.0	0.64	0.0	4.0	23.1
Ortho–phosphate–phosphorus	mg/l	83	8.5	4.03	2.6	19.6	-10.0

Note: n, number of tested samples; SD, standard deviation; Min, minimum; Max, maximum; Rem., removal; and NTU, nephelometric turbidity unit.

Table 5.5. Control (C) outflow parameters for tap water (TW) after two and seven days of contact times.

Parameter	Unit	n	Mean	SD	Min	Max
2–day outflow (TW)–C1						
pH	–	85	6.7	0.39	6.0	7.7
Redox potential	mV	85	42.2	16.50	-7.7	70.8
Turbidity	NTU	85	9.3	6.61	2.0	39.3
Total suspended solids	mg/l	85	14.3	8.16	0.0	35.0
Electrical conductivity	µS/cm	85	84.4	12.15	57.2	117.7
Dissolved oxygen	mg/l	85	9.0	0.87	7.2	11.8
Colour	Pa/Co	85	44.3	30.56	6.0	168.0
Temperature	°C	85	16.5	3.76	5.9	22.4
Biochemical oxygen demand	mg/l	85	7.3	3.45	0.0	18.0
Chemical oxygen demand	mg/l	85	15.9	7.74	4.9	42.8
Ammonia–nitrogen	mg/l	85	0.1	0.12	0.0	0.6
Nitrate–nitrogen	mg/l	85	1.1	0.75	0.1	3.8
Ortho–phosphate–phosphorus	mg/l	85	2.8	1.82	0.9	10.6
2–day outflow (TW)–C2						
pH	–	85	7.4	0.60	6.2	9.3
Redox potential	mV	85	9.6	28.10	-72.2	78.4
Turbidity	NTU	85	4.2	4.37	1.2	32.5
Total suspended solids	mg/l	85	3.9	2.93	0.0	15.0
Electrical conductivity	µS/cm	85	81.5	9.94	57.2	116.2
Dissolved oxygen	mg/l	85	10.4	0.70	8.9	12.0
Colour	Pa/Co	85	8.6	7.66	0.0	34.0
Temperature	°C	85	16.8	4.04	6.0	22.8
Biochemical oxygen demand	mg/l	85	5.4	4.03	0.0	18.0
Chemical oxygen demand	mg/l	85	6.3	2.84	1.0	14.3
Ammonia–nitrogen	mg/l	85	0.1	0.14	0.0	0.6
Nitrate–nitrogen	mg/l	85	0.8	0.53	0.0	3.6
Ortho–phosphate–phosphorus	mg/l	85	2.4	0.63	0.9	4.3
7–day outflow (TW)–C3						
pH	–	83	6.6	0.39	6.0	7.3
Redox potential	mV	83	44.1	17.06	6.1	76.4
Turbidity	NTU	83	12.7	12.56	1.5	69.9
Total suspended solids	mg/l	83	17.8	13.69	0.0	67.0
Electrical conductivity	µS/cm	83	92.9	27.28	60.7	167.2
Dissolved oxygen	mg/l	83	8.9	1.09	5.7	11.2
Colour	Pa/Co	83	56.1	31.45	11.0	143.0
Temperature	°C	83	15.1	4.20	6.2	22.2
Biochemical oxygen demand	mg/l	83	9.1	5.05	0.0	22.0
Chemical oxygen demand	mg/l	83	17.6	6.74	6.3	35.2
Ammonia–nitrogen	mg/l	83	0.1	0.04	0.0	0.2
Nitrate–nitrogen	mg/l	83	0.9	0.42	0.0	2.8
Ortho–phosphate–phosphorus	mg/l	83	3.4	1.47	0.4	7.4
7–day outflow (TW)–C4						
pH	–	83	7.1	0.52	5.9	8.9
Redox potential	mV	83	25.1	24.68	-60.9	89.8
Turbidity	NTU	83	3.7	3.47	1.3	30.1
Total suspended solids	mg/l	83	4.3	5.79	0.0	38.0
Electrical conductivity	µS/cm	83	87.1	20.83	58.4	163.2
Dissolved oxygen	mg/l	83	10.8	1.07	8.4	13.2
Colour	Pa/Co	83	12.7	9.73	0.0	42.0
Temperature	°C	83	15.5	4.17	7.2	22.0
Biochemical oxygen demand	mg/l	83	6.7	4.65	0.0	24.0
Chemical oxygen demand	mg/l	83	7.0	2.48	1.6	17.3
Ammonia–nitrogen	mg/l	83	0.1	0.05	0.0	0.2
Nitrate–nitrogen	mg/l	83	0.8	0.54	0.0	3.0
Ortho–phosphate–phosphorus	mg/l	83	2.4	0.86	0.5	4.6

Note: n, number of tested samples; SD, standard deviation; Min, minimum; Max, maximum; and NTU, nephelometric turbidity unit.

Similarly, removals for wetlands F5 and F7 (HC– and LC–SGW, respectively, and both planted and with seven days of contact time) were compared with those for wetlands F6 and F8 (same design set–up but without plants). Regarding treatment time, the effects of increasing contact time were evaluated by comparing the removals in wetlands (F1, F2, F3 and F4) for two days of treatment contact time with the related removals in wetlands for seven days of treatment time (F5, F6, F7 and F8). Finally, the performance of FTWs under different pollutant loads was tested by comparing the removals in wetlands treating HC–SGW (i.e. F1, F2, F5 and F6) with those treating LC–SGW (i.e. F3, F4, F7 and F8).

5.3.1. Oxygen Demands

Table 5.6 shows that the outflow BOD_5 concentrations were significantly lower than those for both inflow greywater load strengths (HC–SGW and LC–SGW) in all either vegetated or unvegetated wetlands (Figure 5.1).

Table 5.6. Overview of the statistical analysis of variances between inflow and outflow water qualities for different wetlands (F) treating wastewater with high concentrations (HC) and low concentrations (LC) using the Kruskal–Wallis test.

Parameters	Unit	p–values linked to the Kruskal–Wallis tests									
		HC inflow, F1, F2, F5, and F6	*HC inflow and outflow of*				LC inflow, F3, F4, F7, and F8	*LC inflow and outflow of*			
			F1	F2	F5	F6		F3	F4	F7	F8
pH	–	<0.001	0.001	0.395	<0.001	0.140	<0.001	1.000	<0.001	1.000	<0.001
Redox potential	mV	<0.001	0.002	0.086	0.001	0.160	<0.001	1.000	<0.001	1.000	<0.001
Turbidity	NTU[a]	<0.001	1.000	1.000	<0.001	<0.001	<0.001	1.000	0.551	0.107	0.001
Total suspended solids	mg/l	<0.001	1.000	1.000	0.869	1.000	<0.001	1.000	0.009	<0.001	<0.001
Electrical conductivity	µS/cm	<0.001	1.000	1.000	1.000	1.000	<0.001	1.000	0.095	1.000	0.665
Dissolved oxygen	mg/l	<0.001	<0.001	1.000	<0.001	1.000	<0.001	<0.001	1.000	<0.001	0.265
Colour	Pa/Co	<0.001	1.000	1.000	1.000	1.000	<0.001	0.013	<0.001	<0.001	<0.001
Temperature	°C	0.346	na	na	na	na	na	0.003	1.000	0.578	0.179	0.007
Biochemical oxygen demand	mg/l	<0.001	<0.001	<0.001	0.001	<0.001	<0.001	<0.001	<0.001	1.000	<0.001
Chemical oxygen demand	mg/l	<0.001	<0.001	0.013	<0.001	<0.001	<0.001	1.000	1.000	0.897	<0.001
Ammonia–nitrogen	mg/l	<0.001	1.000	1.000	0.006	1.000	<0.001	0.272	<0.001	0.118	<0.001
Nitrate–nitrogen	mg/l	<0.001	<0.001	1.000	1.000	1.000	<0.001	0.134	1.000	1.000	1.000
Ortho–phosphate–phosphorus	mg/l	<0.001	0.266	<0.001	<0.001	<0.001	<0.001	1.000	0.161	0.004	1.000

Note: na, not applicable; NTU, nephelometric turbidity unit.

Although a standard nitrification inhibitor was utilised in determining BOD_5 (APHA, 2005), considerable BOD_5 outflow fluctuations may indicate the presence of nitrification processes during the incubator period (Tanner et al., 1995).

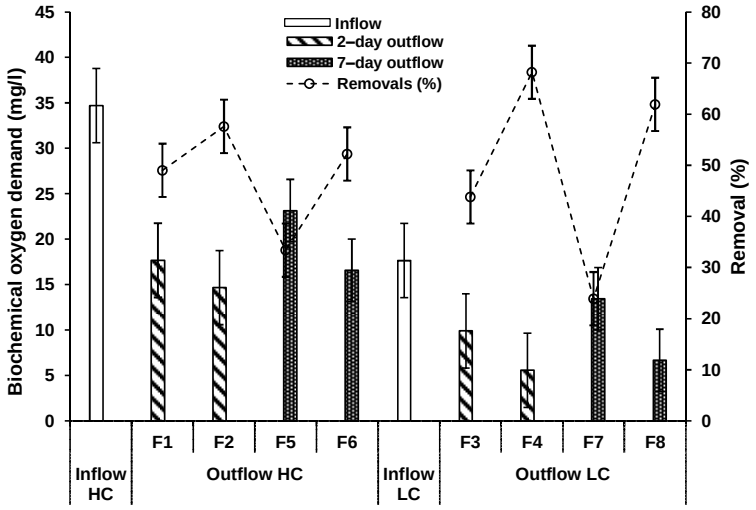


Figure 5.1. Comparison between inflow and outflow synthetic greywater treated in various experimental set–up designs of floating treatment wetlands in terms of biochemical oxygen demand.

The presence of *P. australis* significantly impacted on the increase of BOD_5 concentrations in the outflow of wetlands treating both types of SGW (HC for F1and F5; LC for F3 and F7) compared with those concentrations of unvegetated wetlands (HC for F2 and F6; LC for F4 and F8) for both treatment times (Table 5.7 and Figure 5.1). Tanner et al. (1995) reported that the removal of BOD_5 has not been affected by the presence of vegetation in experimental wetlands. However, Burgoon et al. (1989) reported that BOD_5 removal for unplanted mesocosms has been lower by around 15%.

For HC–SGW, a comparison between vegetated wetlands at 2–day contact time (F1) with those at 7–day contact time (F5) has shown a significant ($p < 0.05$) increase in BOD_5 with increase in contact time. However, there was no significant ($p > 0.05$) effect of treatment time on BOD_5 in wetlands without macrophytes (F2 compared to F6) as shown in Table 5.7. The same trend regarding treatment time effects has been

demonstrated for LC–SGW when comparing F7 with F3 for vegetated wetlands and F8 with F4 for unvegetated systems (Figure 5.1). In this context, Tanner et al. (1995) argued that with increasing retention time of treatment in constructed wetlands, removals of BOD_5 increased by a rate of 16% and 25% for planted and unplanted wetlands, respectively.

In addition, finding showed that the lowest concentrations of outflow BOD_5 were recorded for unvegetated wetlands with 2–day contact times for HC–SGW (with rank relationships F2 < F6 < F1 < F5) and LC–SGW (with rank relationships F4 < F8 < F3 < F7), as shown in Figure 5.1. The behaviour of FTW under vegetated conditions and the effect of contact time can be assessed by studying the trend of BOD_5 changes in control wetlands, which contained tap water. The concentrations of BOD_5 in the vegetated control wetlands C1 (2–day contact time) and C3 (7–day contact time) were significantly ($p < 0.05$) higher than those in the unvegetated wetlands C2 (2–day contact time) and C4 (7–day contact time), respectively, as shown in Tables 5.5 and 5.7.

Furthermore, increasing treatment times significantly increased the BOD_5 concentration in vegetated wetlands (C1 compared to C3). However, there was no significant ($p > 0.05$) effect of increasing treatment time on BOD_5 concentrations in unvegetated wetlands (C2 compared to C4).

For both types of greywater, the increase of treatment time in vegetated floating wetlands reduced their performance in removing BOD_5, which is indirectly required for the biological degradation of organic matter by microorganisms (Gross et al., 2007b; Vymazal and Kröpfelová, 2009).

Table 5.7. Statistical analysis for comparison between water parameter values affected by experimental operational variables in various designs of floating treatment wetlands.

Effects of vegetation on wetlands (F)

		High concentration greywater comparisons						**Low concentration greywater comparisons**					
		2–day contact time F1 & F2			***7–day contact time*** F5 & F6			***2–day contact time*** F3 & F4			***7–day contact time*** F7 & F8		
Parameter	**Unit**	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (value)
pH	–	<0.001	Mann–Whitney	0.146	<0.001	Mann–Whitney	0.141	<0.001	Mann–Whitney	<0.001	<0.001	Mann–Whitney	<0.001
Redox potential	mV	<0.001	Mann–Whitney	0.316	<0.001	Mann–Whitney	0.134	<0.001	Mann–Whitney	<0.001	<0.001	Mann–Whitney	<0.001
Turbidity	NTU	0.257	T–test	0.059	0.002	Mann–Whitney	0.014	<0.001	Mann–Whitney	0.167	<0.001	Mann–Whitney	0.304
Total suspended solids	mg/l	0.002	Mann–Whitney	0.131	0.002	Mann–Whitney	0.010	<0.001	Mann–Whitney	0.090	<0.001	Mann–Whitney	0.424
Electrical conductivity	µS/cm	0.201	T–test	0.178	<0.001	Mann–Whitney	0.021	<0.001	Mann–Whitney	0.009	<0.001	Mann–Whitney	0.002
Dissolved oxygen	mg/l	<0.001	Mann–Whitney	<0.001	0.092	T–test	<0.001	0.458	T–test	<0.001	0.105	T–test	<0.001
Colour	Pa/Co	0.063	T–test	0.72	0.336	T–test	0.029	<0.001	Mann–Whitney	0.551	<0.001	Mann–Whitney	0.408
Temperature	°C	<0.001	Mann–Whitney	0.561	<0.001	Mann–Whitney	0.570	<0.001	Mann–Whitney	0.027	<0.001	Mann–Whitney	0.347
Biochemical oxygen demand	mg/l	0.008	Mann–Whitney	0.003	<0.001	Mann–Whitney	<0.001	<0.001	Mann–Whitney	<0.001	<0.001	Mann–Whitney	<0.001
Chemical oxygen demand	mg/l	0.303	T–test	0.016	<0.001	Mann–Whitney	0.018	<0.001	Mann–Whitney	0.017	<0.001	Mann–Whitney	<0.001
Ammonia–nitrogen	mg/l	<0.001	Mann–Whitney	0.212	<0.001	Mann–Whitney	<0.001	<0.001	Mann–Whitney	0.086	<0.001	Mann–Whitney	1.000
Nitrate–nitrogen	mg/l	0.003	Mann–Whitney	<0.001	<0.001	Mann–Whitney	0.006	<0.001	Mann–Whitney	0.002	<0.001	Mann–Whitney	0.003
Ortho–phosphate–phosphorus	mg/l	0.002	Mann–Whitney	0.002	0.006	Mann–Whitney	0.013	<0.001	Mann–Whitney	0.054	<0.001	Mann–Whitney	<0.001

Note: p value, significant difference, if $p < 0.05$ and not significant if $p > 0.05$; M–W, Mann–Whitney U–test; and NTU, nephelometric turbidity unit.

Table 5.7. (Continued).

Effects of increasing contact time on wetlands (F)

		High concentration greywater comparisons						**Low concentration greywater comparisons**					
		With vegetation F1 & F5			***Without vegetation*** F2 & F6			***With vegetation*** F3 & F7			***Without vegetation*** F4 & F8		
Parameter	**Unit**	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (value)
pH	–	<0.001	Mann–Whitney	0.886	<0.001	Mann–Whitney	0.751	<0.001	Mann–Whitney	0.575	<0.001	Mann–Whitney	0.805
Redox potential	mV	<0.001	Mann–Whitney	0.870	<0.001	Mann–Whitney	1.000	<0.001	Mann–Whitney	0.655	<0.001	Mann–Whitney	0.797
Turbidity	NTU	0.001	Mann–Whitney	0.093	0.169	T–test	0.405	<0.001	Mann–Whitney	0.048	<0.001	Mann–Whitney	0.030
Total suspended solids	mg/l	<0.001	Mann–Whitney	0.084	0.189	T–test	0.037	<0.001	Mann–Whitney	0.007	<0.001	Mann–Whitney	0.009
Electrical conductivity	µS/cm	<0.001	Mann–Whitney	0.149	<0.001	Mann–Whitney	0.492	<0.001	Mann–Whitney	0.041	<0.001	Mann–Whitney	0.533
Dissolved oxygen	mg/l	0.003	Mann–Whitney	0.113	0.102	T–test	0.022	0.419	T–test	0.876	0.082	T–test	0.003
Colour	Pa/Co	0.051	Mann–Whitney	0.358	0.041	Mann–Whitney	0.104	<0.001	Mann–Whitney	0.084	0.274	T–test	0.063
Temperature	°C	<0.001	Mann–Whitney	0.158	<0.001	Mann–Whitney	0.099	<0.001	Mann–Whitney	0.018	<0.001	Mann–Whitney	0.112
Biochemical oxygen demand	mg/l	<0.001	Mann–Whitney	<0.001	0.002	Mann–Whitney	0.122	0.001	Mann–Whitney	<0.001	<0.001	Mann–Whitney	0.264
Chemical oxygen demand	mg/l	0.006	Mann–Whitney	0.910	<0.001	Mann–Whitney	0.214	<0.001	Mann–Whitney	0.821	0.132	T–test	<0.001
Ammonia–nitrogen	mg/l	0.004	Mann–Whitney	0.002	<0.001	Mann–Whitney	0.033	<0.001	Mann–Whitney	0.690	<0.001	Mann–Whitney	0.175
Nitrate–nitrogen	mg/l	<0.001	Mann–Whitney	0.001	<0.001	Mann–Whitney	0.005	<0.001	Mann–Whitney	0.008	<0.001	Mann–Whitney	0.028
Ortho–phosphate–phosphorus	mg/l	0.020	Mann–Whitney	0.053	0.048	Mann–Whitney	0.025	<0.001	Mann–Whitney	<0.001	<0.001	Mann–Whitney	0.001

Note: p value, significant difference, if $p < 0.05$ and not significant if $p > 0.05$; M–W, Mann–Whitney U–test; and NTU, nephelometric turbidity unit.

Table 5.7. (Continued).

Effects of vegetation and increasing contact time on control (C) floating wetlands

		Effect of vegetation						**Effect of increasing contact time**					
		2–day contact time C1 & C2			***7–day contact time*** C3 & C4			***With vegetation*** C1 & C3			***Without vegetation*** C2 & C4		
Parameter	**Unit**	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
pH	–	<0.001	Mann–Whitney	<0.001	0.003	Mann–Whitney	<0.001	0.004	Mann–Whitney	0.540	0.013	Mann–Whitney	<0.001
Redox potential	mV	<0.001	Mann–Whitney	<0.001	0.044	Mann–Whitney	<0.001	0.004	Mann–Whitney	0.563	0.006	Mann–Whitney	0.001
Turbidity	NTU	<0.001	Mann–Whitney	<0.001	<0.001	Mann–Whitney	<0.001	<0.001	Mann–Whitney	0.320	<0.001	Mann–Whitney	0.065
Total suspended solids	mg/l	<0.001	Mann–Whitney	<0.001	<0.001	Mann–Whitney	<0.001	<0.001	Mann–Whitney	0.275	<0.001	Mann–Whitney	0.181
Electrical conductivity	µS/cm	<0.001	Mann–Whitney	0.256	<0.001	Mann–Whitney	0.549	<0.001	Mann–Whitney	0.353	<0.001	Mann–Whitney	0.035
Dissolved oxygen	mg/l	0.165	T–test	<0.001	0.559	T–test	<0.001	0.425	T–test	0.595	0.016	Mann–Whitney	0.006
Colour	Pa/Co	<0.001	Mann–Whitney	<0.001	<0.001	Mann–Whitney	<0.001	<0.001	Mann–Whitney	0.006	<0.001	Mann–Whitney	0.002
Temperature	°C	<0.001	Mann–Whitney	0.303	<0.001	Mann–Whitney	0.524	<0.001	Mann–Whitney	0.018	<0.001	Mann–Whitney	0.027
Biochemical oxygen demand	mg/l	<0.001	Mann–Whitney	<0.001	<0.001	Mann–Whitney	0.001	<0.001	Mann–Whitney	0.008	<0.001	Mann–Whitney	0.051
Chemical oxygen demand	mg/l	<0.001	Mann–Whitney	<0.001	<0.001	Mann–Whitney	<0.001	<0.001	Mann–Whitney	0.004	0.007	Mann–Whitney	0.143
Ammonia–nitrogen	mg/l	<0.001	Mann–Whitney	0.939	<0.001	Mann–Whitney	0.080	<0.001	Mann–Whitney	<0.001	<0.001	Mann–Whitney	<0.001
Nitrate–nitrogen	mg/l	<0.001	Mann–Whitney	0.017	<0.001	Mann–Whitney	0.092	<0.001	Mann–Whitney	0.109	<0.001	Mann–Whitney	0.637
Ortho–phosphate–phosphorus	mg/l	<0.001	Mann–Whitney	0.637	<0.001	Mann–Whitney	<0.001	<0.001	Mann–Whitney	<0.001	<0.001	Mann–Whitney	0.686

Note: p value, significant difference, if $p < 0.05$ and not significant if $p > 0.05$; M–W, Mann–Whitney U–test; and NTU, nephelometric turbidity unit.

Table 5.8 shows the effect of contamination strength on the performance of FTW when using vegetated and unvegetated systems with different contact times. The statistical analysis using the Mann–Whitney test showed that there is no significant ($p > 0.05$) effect of greywater strength on BOD_5 removals when comparing the vegetated wetlands of F1 with F3 (2–day contact time) and F5 with F7 (7–day contact time). However, BOD_5 has been removed significantly in unvegetated wetlands F4 and F8 (treated LC–SGW) compared to those of F2 and F6 (HC–SGW). Furthermore, high removal of BOD_5 was indicated by 68.2% in unvegetated wetlands treating LC–SGW for a 2–day contact time. Wetland removals followed the rank relationship F4 > F8 > F2 > F6 > F1 > F3 > F5 > F7, as shown in Figure 5.1.

Table 5.8. Statistical analysis using the Mann–Whitney U–test for comparison of water quality parameter removals in floating wetlands treating different pollutant loads.

Removal (%)	Unit	Vegetated wetlands		Unvegetated wetlands	
		2–day contact time	*7–day contact time*	*2–day contact time*	*7–day contact time*
		F1 & F3	F5 & F7	F2 & F4	F6 & F8
Turbidity	NTU	0.935	0.929	0.006	<0.001
Total suspended solids	mg/l	0.233	0.063	<0.001	<0.001
Dissolved oxygen	mg/l	0.128	0.029	0.069	0.013
Colour	Pa/Co	0.004	0.059	<0.001	<0.001
Biochemical oxygen demand	mg/l	0.572	0.095	0.010	0.031
Chemical oxygen demand	mg/l	<0.001	<0.001	0.032	<0.001
Ammonia–nitrogen	mg/l	0.010	<0.001	0.001	<0.001
Nitrate–nitrogen	mg/l	0.065	0.882	0.593	0.590
Ortho–phosphate–phosphorus	mg/l	0.845	<0.001	0.664	0.001

Note: NTU, nephelometric turbidity unit.

A comparison of the outflow COD concentrations for HC–SGW with those of the inflow have shown significant decreases in all vegetated and unvegetated wetlands for both contact time of treatment. In contrast, the COD concentrations of the LC–SGW outflow did not show any significant change within two days of treatment time, while it increased slightly for the vegetated wetlands F3 and dropped for the unvegetated wetlands F4 compared to the LC–SGW inflow values (Figure 5.2). Furthermore, a

significant ($p < 0.05$) increase has been noted for the outflow COD concentrations of vegetated wetlands F7 after seven days of treatment, and a significant ($p < 0.05$) reduction in COD levels for unvegetated wetlands F8, which treated LC–SGW at the same contact time, was recorded (Tables 5.3 and 5.4).

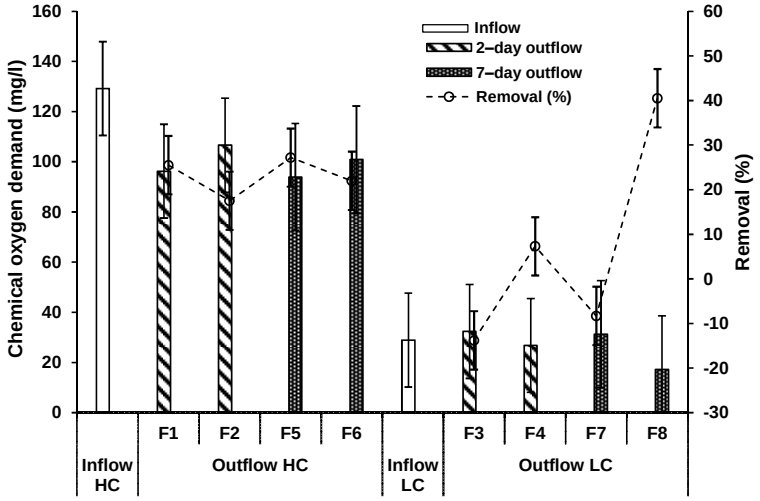


Figure 5.2. Comparison between inflow and outflow synthetic greywater treated in various experimental set–up designs of floating treatment wetlands in terms of chemical oxygen demand.

The presence of *P. australis* in wetlands F1 (2–day contact time) and F5 (7–day contact time) treating HC–SGW has significantly ($p < 0.05$) improved the reduction of COD concentrations compared to those of unvegetated wetlands F2 (2–day contact time) and F6 (7–day contact time) as indicated in Figure 5.2. In contrast, the presence of macrophytes in wetlands treating LC–SGW has significantly ($p < 0.05$) increased the COD concentrations in wetlands F3 compared to the unvegetated wetlands F4 (2–day contact time), and F7 (with vegetation) compared to F8 (without vegetation) for 7–day contact time (Table 5.7). This confirms findings reported by Akratos and Tsihrintzis (2007), indicating that the COD removal in horizontal–flow constructed wetland systems was significantly improved when using plants (*Typha latifolia* in this case) compared to those systems, which were unplanted.

In addition, increasing the contact time led to a significant ($p < 0.05$) effect in reduction of the COD concentrations of unvegetated wetlands F8, which treated LC–SGW compared to the outflow COD for F4 (2–day contact time). Slight COD reductions were noted for other floating wetlands (Table 5.7). In general, the mean outflow in terms of COD concentrations for HC–SGW was ranked as follows: F5 < F1 < F6 < F2. In comparison, for LC–SGW, the ranking was F8 < F4< F7 < F3 (Figure 5.2).

However, the control floating wetlands (containing tap water) have shown that COD concentrations at the present of *P. australis* in wetlands C1 (2–day contact time) and C3 (7–day contact time) were significantly greater than those of unvegetated wetlands C2 (2–day contact time) and C4 (7–day contact time). Furthermore, an increase of contact time led to an elevated outflow COD concentration, which was significant, if *P. australis* was present (C1 compared with C3). However, there was no significant change between outflow COD concentrations of unvegetated wetlands C2 and C4 with increasing contact time of treatment (Tables 5.5 and 5.7).

The effect of pollutant loads can be distinguished by comparing COD removal for different design set–ups of floating wetlands treating HC–SGW and LC–SGW. For all vegetated and unvegetated floating wetlands, the COD removals from HC–SGW (F1, F2, F5 and F6) were significantly higher than those removals for wetlands treating LC–SGW (F3, F4, F7 and F8) for both contact times of treatment (Table 5.8). In general, the COD removal values in FTW for different set–up operational variables followed the order: F8 > F5 > F1 > F6 > F2 > F4 > F7 > F3 (Figure 5.2).

Correlation analysis results indicated that BOD_5 was significantly positively correlated with COD ($r = 0.306$, $p = 0.005$) for HC–SGW inflow. However, there was no correlation between BOD_5 and COD for LC inflow greywater. For HC–SGW outflow,

significantly ($p < 0.01$) negative correlations (($r = -0.311$, $p = 0.004$) and ($r = -0.315$, $p = 0.003$)) were calculated between BOD_5 and COD for vegetated wetlands F1 and unvegetated wetlands F2, respectively, at 2–day contact time.

However, after seven days of treatment, for the same design but for wetlands F5 and F6, the correlations became significantly ($p < 0.01$) positive (($r = 0.400$, $p = 0.000$) and ($r = 0.398$, $p = 0.000$), respectively). The correlation analysis of wetlands treating LC–SGW has shown no correlation between the outflow BOD_5 and COD for 2–day contact time concerning wetlands F3 (vegetated) and F4 (unvegetated). In contrast, BOD_5 correlated significantly ($p < 0.01$) positively ($r = 0.299$, $p = 0.006$) with COD for the vegetated wetlands F7 only after seven days of treatment. In general, the overall behaviour of floating treatment systems has shown a significantly ($p < 0.01$) positive correlation between BOD_5 and COD ($r = 0.838$, $p = 0.000$). Moreover, the reduction in BOD_5 and COD concentrations of aquatic systems mainly depends upon the oxygen concentration in the outflow as decomposition of pollutants occurs through oxidation reactions (Vymazal, 2010).

The biodegradability of greywater can be measured by the ratio between BOD_5 and COD. In this study, the BOD_5 to COD ratio of inflow SGW before utilisation in the experiment was 0.27 and 0.61 for HC–SGW and LC–SGW, respectively (Figure 5.3). After two days of treatment by vegetated floating wetlands, the ratio decreased to 0.18 in wetlands F1 (treated HC–SGW) and to 0.31 in wetlands F3 (treated LC–SGW), compared with a ratio of 0.46 for control wetlands C1 (containing tap water). While the ratio for unvegetated wetlands for the same contact time was 0.14 for F2 (treated HC–SGW) and 0.21 for F4 (treated LC–SGW), a ratio of 0.86 was calculated for the control wetlands C2 (containing tap water).

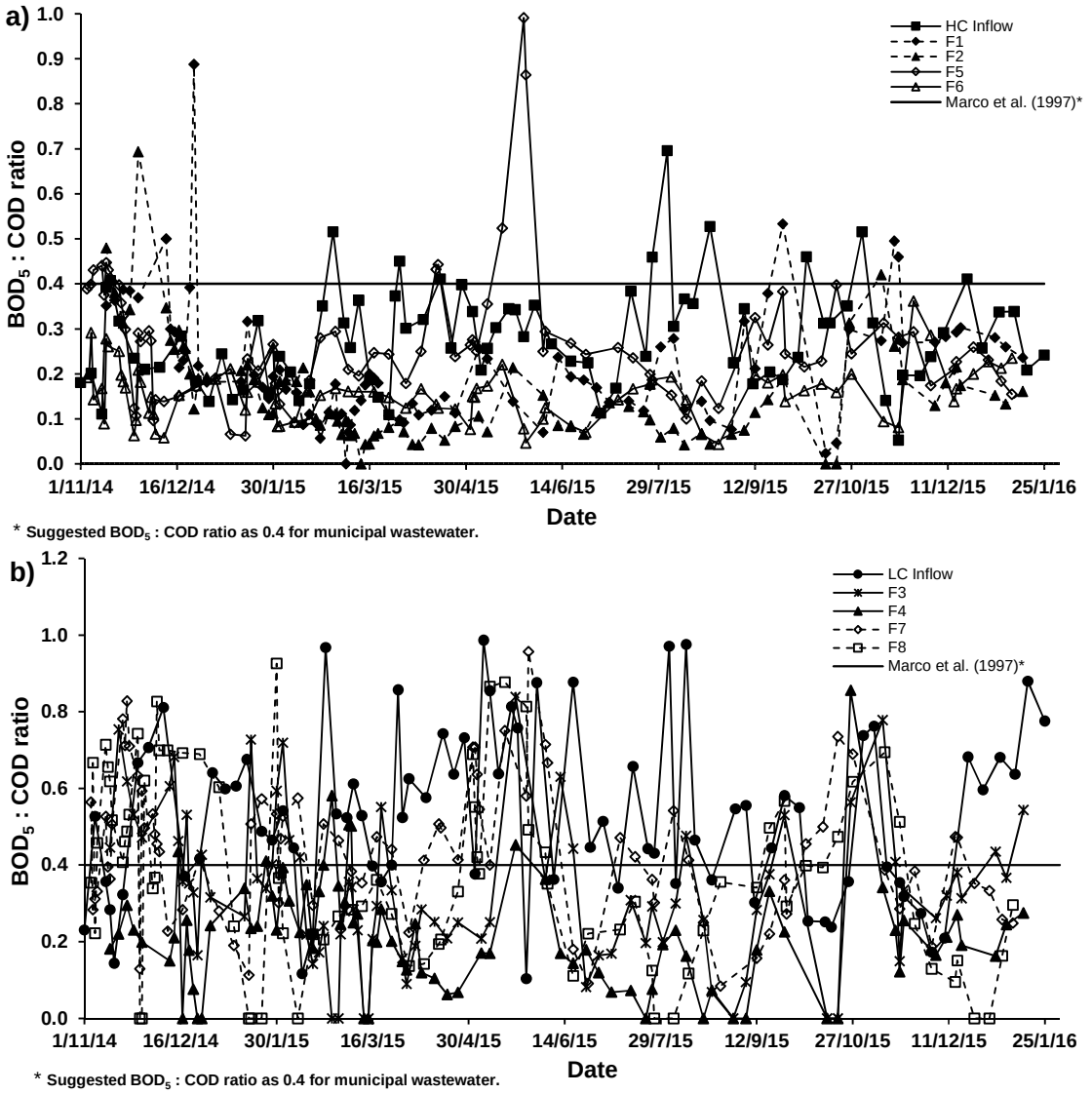


Figure 5.3. Comparison of the biodegradation ratio (BOD_5 : COD) for the system inflow and outflow regarding: (a) a high contamination load (HC–SGW); and (b) a low contamination load (LC–SGW).

After seven days of treatment, the BOD_5 to COD ratio of the outflow from the vegetated wetlands was 0.25 for F5 (treated HC–SGW) and 0.43 for wetlands F7 (treated LC–SGW), compared to a ratio of 0.52 for control wetlands C3 (containing tap water). For unvegetated wetlands, the ratio was 0.16 for wetlands F6 (treated HC–SGW) and 0.39 for wetlands F8 (treated LC–SGW), compared with a ratio of 0.96 for control wetlands C4 (contained tap water). Similarly, low biodegradability of greywater has been

reported by Jefferson et al. (1999). However, after two hours of treatment, the BOD_5 to COD ratio of the greywater increased to 0.41, which is comparable with municipal wastewater, where the ratio is often between 0.4 and 0.8 (Marco et al., 1997). This demonstrates that the proportion of biodegradable contaminants in the treated water was higher than that in the raw greywater (Figure 5.3).

5.3.2. Nutrients

A comparative statistical analysis between the inflow greywater characteristics with their corresponding ones for outflow is shown in Table 5.6. In terms of NH_4–N, the concentrations did not change after two days of treatment for HC–SGW in the vegetated and unvegetated wetlands F1 and F2, respectively, compared to inflow concentrations. For the 7–day contact time, NH_4–N concentrations increased significantly ($p < 0.05$) in vegetated wetlands F5 (treated HC–SGW) compared to those of the inflow. However, no significant ($p > 0.05$) difference in concentration of the unvegetated wetlands F6 was noted. Furthermore, there were significant differences in outflow concentrations of NH_4–N in all vegetated and unvegetated wetlands, which treated LC–SGW (Figure 5.4).

In comparison, the presence of macrophytes has not affected the NH_4–N concentrations of the outflow HC–SGW for wetlands F1 and F2 (2–day contact times). However, the NH_4–N concentration increased significantly ($p <0.05$) in vegetated wetlands F5 (7–day contact time) compared to those concentrations of unvegetated wetlands F6 (7–day contact time). In addition, no significant ($p > 0.05$) effect of macrophytes on the concentrations of NH_4–N in floating wetlands treating LC–SGW at both contact times of treatment compared to the NH_4–N concentrations of unvegetated wetlands were noted (Table 5.7).

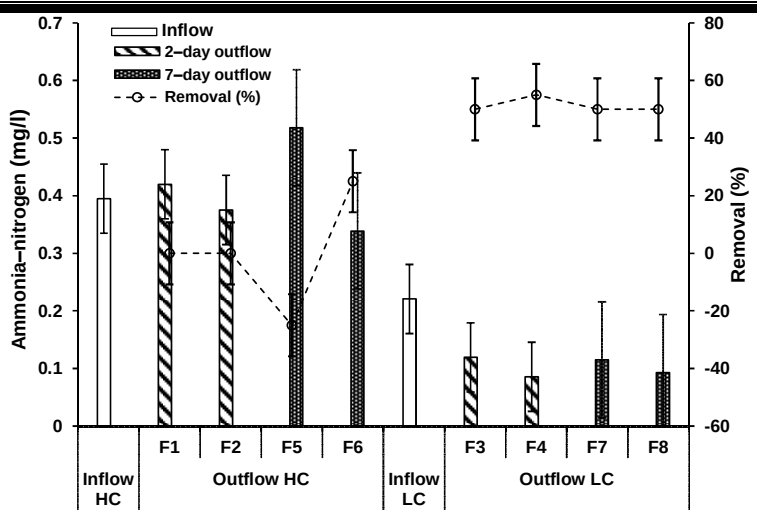


Figure 5.4. Comparison between inflow and outflow synthetic greywater treated in various experimental set–up designs of floating treatment wetlands in terms of ammonia–nitrogen.

Increasing contact times have insignificantly ($p > 0.05$) affected NH_4–N concentrations in the outflow of all wetlands treating LC–SGW. In contrast, increasing contact times at the presence of macrophytes in wetlands F5 (treated HC–SGW) caused a significant ($p < 0.05$) increase in NH_4–N concentrations, compared to those concentrations in the outflow of vegetated wetlands F1 (2–day contact time). A significant ($p < 0.05$) reduction in NH_4–N concentration was calculated for the outflow of unvegetated wetlands F6 treating HC–SGW for seven days, compared to unvegetated wetlands F2 (2–day contact time) as shown in Table 5.7. The mean values of NH_4–N concentrations of floating wetlands ranked as follows: F6 < F1 = F2 < F5 (treated HC–SGW); and F4 < F3 = F7 = F8 (treated LC–SGW) (Figure 5.4).

The effect of macrophyte presence on outflow NH_4–N concentrations for the control wetlands, which contained tap water, was similar to those of wetlands treating LC–SGW, if vegetated wetlands are compared to unvegetated ones. There was no significant ($p > 0.05$) effect of vegetation on NH_4–N concentrations, if systems C1 are compared with C2 (2–day contact time), and systems C3 with C4 (7–day contact time).

The outflow removals of NH_4–N for wetlands treating LC–SGW were significantly ($p < 0.05$) higher than those removals of corresponding wetlands treating HC–SGW for two days of contact time (vegetated wetlands F1 compared with F3 and unvegetated wetlands F2 compared with F4) and seven days of contact time (vegetated wetlands F5 compared with F7 and unvegetated wetlands F6 compared with F8). In general, the overall outflow removals in terms of NH_4–N were ranked in FTW as follows: F4 > F3 = F7 = F8 > F6 > F1 = F2 > F5 (Figure 5.4, and Tables 5.3 and 5.4). Considering that NH_4–N volatilization and plant uptake are primary removal mechanisms in floating treatment wetland systems (Ferrara and Avci, 1982), the extent of ammonia generation from the reduction of other nitrogenous forms such as NO_3–N and organic–N may have been substantial in accounting for the observed increases in some wetlands (Wallace et al., 2016).

The correlation analysis indicated that the mean NH_4–N inflow concentration was significantly ($p < 0.01$) positively correlated ($r = 0.434$, $p = 0.000$) with COD of the LC–SGW inflow, while no correlation was observed between NH_4–N and BOD_5. Although NH_4–N did not correlate with both BOD_5 and COD in HC–SGW inflow, NH_4–N outflow for vegetated floating wetlands F1 (2–day contact time) and F5 (7–day contact time) correlated significantly ($p < 0.01$) positively (($r = 0.279$, $p = 0.010$) and ($r = 0.605$, $p = 0.000$), respectively) with COD. However, a significant ($p < 0.01$) positive correlation ($r = 0.609$, $p = 0.000$) with BOD_5 was shown for F5. For treated HC–SGW and the unvegetated F6 (7–day contact time), NH_4–N was significantly ($p < 0.05$) positively correlated ($r = 0.219$, $p = 0.047$) with COD, while no correlation with BOD_5 has been noted. Moreover, these correlation relationships were not found for F2 (2–day contact time).

For LC–SGW, NH_4–N of F3 (vegetated; 2–day contact time), F4 (unvegetated; 2–day contact time) and F7 (vegetated; 7–day contact time) were significantly ($p < 0.05$) negatively correlated (($r = -0.220$, $p = 0.043$), ($r = -0.273$, $p = 0.012$) and ($r = -0.262$, $p = 0.017$), respectively) with COD. However, NH_4–N of F8 (unvegetated; 7–day contact time) was significantly ($p < 0.05$) positively correlated ($r = 0.250$, $p = 0.023$) with BOD_5. For the control floating wetlands (containing tap water), NH_4–N of C1 (vegetated; 2–day contact time) was significantly ($p < 0.01$) negatively correlated ($r = -0.282$, $p = 0.009$) with BOD_5. While the NH_4–N outflow of C4 (unvegetated; 7–day contact time) was significantly ($p < 0.05$) negatively correlated (($r = -0.219$, $p = 0.047$) and ($r = -0.229$, $p = 0.037$)) with BOD_5 and COD, respectively.

In general, the overall correlation analysis for NH_4–N outflow values of floating treatment wetland systems indicated significant ($p < 0.01$) positive correlations (($r = 0.876$, $p = 0.000$) and ($r = 0.743$, $p = 0.002$), respectively) with both BOD_5 and COD. The observed increases in concentrations of NH_4–N for some FTW were most likely due to the processes denoted above and are characterised by negative correlations with COD, since no significant ($p > 0.05$) correlation with BOD_5 has been found. For wetlands treating LC–SGW, NH_4–N reductions were due to the positive correlations with both BOD_5 and COD, which is in agreement with other findings (Wallace et al., 2016).

Although floating wetland systems can be used to purify eutrophic water, these technologies are not efficient for removing nitrogenous compounds, particularly NO_3−N, because most greywaters lack biodegradable organic matter, which can be efficiently utilized by denitrifying bacteria. This results commonly in low removal efficiencies regarding nitrogenous compounds (Xiong et al., 2012; Cao et al., 2016).

The outflow NO_3–N concentrations (Tables 5.3–5.5) were statistically compared with those of NO_3–N (Table 5.2) for both types of inflow SGW. Significant ($p < 0.05$) increases in terms of NO_3–N were noted for outflows of vegetated wetlands F1 (HC–SGW) and F3 (LC–SGW) after two days of treatment compared to those of inflow for HC–SGW and LC–SGW, respectively (Table 5.6 and Figure 5.5).

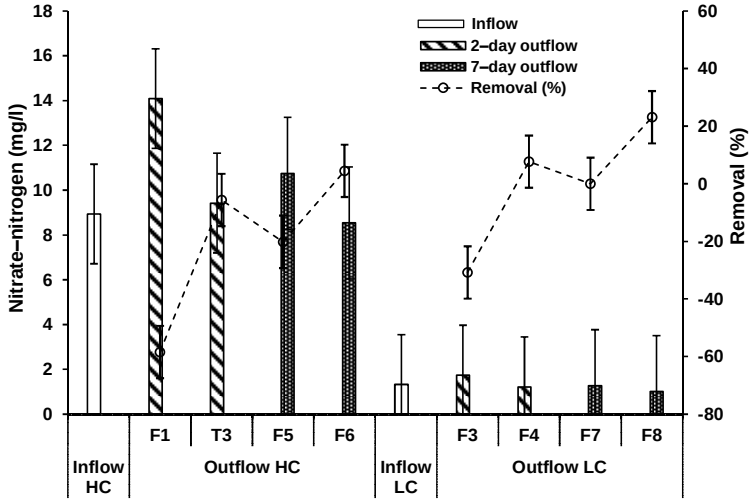


Figure 5.5. Comparison between inflow and outflow synthetic greywater treated in various experimental set–up designs of floating treatment wetlands in terms of nitrate–nitrogen.

The presence of *P. australis* in FTW affected significantly ($p < 0.05$) NO_3–N concentration increases in the outflow compared to those outflow concentrations of unvegetated wetlands for both types of SGW. In contract, increasing contact times affected significantly ($p < 0.05$) decreasing NO_3–N concentrations within the outflow compared to those outflow concentrations of wetlands treating both types of SGW for two days of contact time (Table 5.7).

In general, the mean wetland NO_3–N outflow concentrations for HC–SGW and LC–SGW were ranked as follows: F6 < F2 < F5 < F1, and F8 < F4 < F7 < F3 in that order (Figure 5.5).

The heterogeneity of physical and chemical properties of FTW allows for nitrification–denitrification processes to be applied simultaneously (Hamersley and Howes, 2002). In aerobic conditions, biofilms containing heterotrophic and nitrifying bacteria form on the surfaces of sedimentary and suspended organic matter.

The NO_3–N produced aerobically by nitrification diffuses into these anaerobic microsites, where it becomes available to denitrifying bacteria. As in nature, simultaneous biological nutrient removal (nitrification–denitrification) in wastewater is enhanced by redox potential heterogeneity (Daigger and Littleton, 2000). Studies have shown that greywater is nutrient–deficient, which impacts on the treatment efficiency (Jefferson et al., 2001; 2004).

Therefore, nitrification is not generally considered an important mechanism in FTW due to the lack of nitrifying bacteria in the facultative zone and ready uptake by plants (Ferrara and Avci, 1982), and was not expected to contribute significantly in ammonia conversion (Wallace et al., 2016). Since the roots and rhizomes systems of macrophytes can be damaged by various causes, consequently, an assessment of the removal performance for nitrogenous compounds could be affected (Cao et al., 2016). Once plant tissue degrades, organic nitrogen may be released as algae and bacteria continuously uptake and release nitrogen back to the wetland system (Wallace et al., 2016).

In control wetlands, the only significant effect of macrophytes was linked to NO_3–N in C1 after two days of treatment, if compared to unvegetated wetlands C2. However, no significant difference with increasing contact time of treatment was noticed for NO_3–N concentrations of the outflows of control wetlands (Table 5.7).

Furthermore, no significant ($p > 0.05$) effect of pollutant loads on the performance of FTW in terms of NO_3–N removals was noted (Table 5.8). In general, the outflow removal values of NO_3–N in FTW were ranked as follows: F8 > F4 > F6 > F7 > F2 > F5 > F3 > F1 (Figure 5.5).

All floating wetlands treating HC–SGW for both contact times of treatment showed significant ($p < 0.05$) reductions in the mean concentrations of outflow PO_4–P compared to those of the inflow (Table 5.6). For LC–SGW, no significant ($p > 0.05$) difference in terms of PO_4–P concentrations between outflow and inflow for the vegetated wetlands F3 after two days of treatment were noted. However, unvegetated wetlands F4 showed significant ($p < 0.05$) drops in outflow PO_4–P concentrations (Figure 5.6).

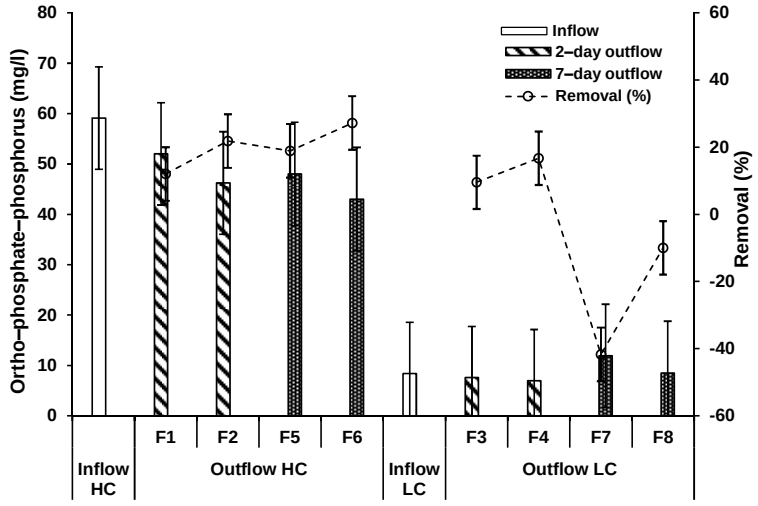


Figure 5.6. Comparison between inflow and outflow synthetic greywater treated in various experimental set–up designs of floating treatment wetlands in terms of ortho–phosphate–phosphorus.

At 7–day contact time, only significant ($p < 0.05$) increases in outflow PO_4–P concentrations were found for vegetated wetlands F7. The presence of macrophytes increased the outflow PO_4–P concentrations in wetlands treating HC–SGW at both treatment times (F1 and F5 compared to F2 and F6, respectively), as indicated in Table

5.7. Vegetation effected significantly PO_4–P concentration increases in wetlands F7 treating LC–SGW compared to those concentrations of unvegetated wetlands F8 at 7–day contact time.

The statistical analysis indicated a significant ($p < 0.05$) decrease in PO_4–P outflows of unvegetated wetlands F6 (treated HC–SGW) compared to those concentrations of wetlands F2 (2–day contact time) when increasing the contact time of treatment. In contrast, an increasing contact time affects significantly ($p < 0.05$) increasing PO_4–P concentrations for both vegetated and unvegetated wetlands F7 and F8 compared to those concentrations of wetlands (2–day contact time) F3 and F4, respectively (Table 5.7). In general, the outflow mean values of PO_4–P concentrations were ranked in wetlands treating HC–SGW and LC–SGW as follows: F6 < F5 < F1 < F2, and F4 < F3 < F8 < F7, respectively (Figure 5.6).

For control floating wetlands, which contained tap water, the presence of vegetation significantly affects increasing PO_4–P concentrations in the outflows of wetlands C3 after seven days of treatment compared to those concentrations of unvegetated wetlands C4. Furthermore, an increase of the contact time of treatment affects significantly increasing PO_4–P concentrations in the vegetated wetlands C3 compared with those concentrations of wetlands C1 (Table 5.7).

The removals of PO_4–P concentrations by wetlands F5 (vegetated) and F6 (unvegetated) treating HC–SGW at 7–day contact time were significantly lower than those concentrations of wetlands F7 (vegetated) and F8 (unvegetated) treating LC–SGW in that order (Table 5.8). In general, the outflow removals in terms of PO_4–P in FTW were ranked as follows: F6 > F2 > F5 > F4 > F1 > F3 > F8 > F7 (Figure 5.6).

As reported, the settling of PO_4–P precipitates is a well–documented process in ponds, wetlands and similar systems (Ann et al., 1999; Peng et al., 2007) and was likely a contributing factor to the PO_4–P reduction throughout the systems. The uptake by algae and/or plants was also a likely factor in the PO_4–P decline. However, this is a seasonal process that would likely have reversed during plant die–off (Wallace et al., 2016). Furthermore, the removal of nitrogen components, which are present as NO_3 and/or NH_4 in the effluent, occurs through natural metabolic processes of plants and associated microbes including ammonium oxidation, nitrification and denitrification. In addition, plants take–up these two entities as nutrients (Vymazal, 2010). Phosphorous is an essential nutrient for both plants and bacteria, although the portion of phosphorus, which is taken–up by plants is significantly lower than nitrogen (Brix, 1994).

The optimisation of hydraulic retention time is necessary for decreasing wastewater treatment time and promoting efficient pollutant removal. In this context, biodegradation and nutrient balancing play vital roles in elevating treatment efficiency of biological systems. Furthermore, adequate trace nutrient concentrations including metal ions and vitamins are required to support organisms present in treatment systems to allow for a diverse community to survive. In this study, the inflow LC–SGW showed an adequate biodegradation rate of 0.61 (in terms of the BOD_5 : COD ratio), which is similar to findings reported by F. Li et al. (2009). In comparison, the ratio was 0.27 for the inflow of HC–SGW (Figure 5.3).

For biological wastewater degradation, a COD : N : P ratio of 100 : 20 : 1 has been suggested by Metcalf et al. (1991) and a ratio of 100 : 10 : 1 has been proposed by Beardsley and Coffey (1985). The HC–SGW and LC–SGW inflows were deficient and unbalanced in terms of nutrients and organic matter (Figure 5.7) due to the exclusion of urine and excreta (Abed and Scholz, 2016). An optimised nitrogen to phosphorus ratio

can make a significant difference to the rates of oxygen uptake and COD removal for both real and SGW (Al–Jayyousi, 2003).

Table 5.9. Comparison of the nutrient balance ratio between the reported, inflow and outflow ratios of various set–up designs of floating treatment wetlands.

	Inflow HC	***Outflow HC***			***Inflow LC***	***Outflow LC***				***Metcalf and Eddy Inc. (1991)****
	(mg/l)									
		F1	**F2**	**F5**	**F6**		**F3**	**F4**	**F7**	**F8**	**(Ratio)**
P	59.1	52.0	46.2	48.0	43.0	8.4	7.6	7.0	11.9	8.5	1
N	9.3	14.5	9.8	11.2	8.8	1.5	1.8	1.29	1.4	1.1	20
COD	129.2	96.3	106.0	94.0	100.8	28.9	32.4	26.8	31.3	17.2	100

*Suggested COD : N : P ratio as 100 : 20 : 1 for wastewater.

According to Jefferson et al. (2001), the deficiency of both macronutrients and trace nutrients in the greywater can limit the treatment efficiency of biological processes. An enhancement of organic degradation in biological treatment systems, which deal with a nutrient–limited wastewater, could be achieved by supplementation of macronutrients. In contrast, Leal et al. (2007) claimed that efficient microorganism populations dominate systems treating greywater with limited nutrients and a BOD_5 to COD ratio of around 0.5. Furthermore, the exclusion of kitchen discharge causes the deficiency of trace nutrients in greywaters, which was reported by Jefferson et al. (2001). F. Li et al. (2009) reported that kitchen greywater does not lack nutrients, and a small amount of kitchen effluent supplement is therefore recommended to maintain an optimal COD : N : P ratio. Furthermore, phosphorus is relatively high in laundry discharge (Eriksson et al., 2002).

5.3.3. Total Suspended Solids and Turbidity

The overall removal efficiencies for TSS and turbidity in floating treatment wetlands were high for both HC–SGW and LC– SGW due to sedimentation, trapping in biofilms and root system, and biodegradation mechanisms. These observations agreed with previous publications (Dunne et al., 2012; Borne et al., 2013). However, in this study, outflow treated SGW was agitated before sampling to simulate the washing–out of solids (Zhang et al., 2015) and to encourage the possibility of solids to dissolve and/or re–suspend into the water column (Stewart et al., 2008; Van de Moortel et al., 2010; Tanner and Headley, 2011). Agitation allowed for the study of macrophyte rhizomes in removing solids.

In comparison with inflow mean values, only significant ($p < 0.05$) decreases were noted for outflow TSS concentrations of vegetated wetlands treating HC–SGW at 7–day contact time. Both vegetated and unvegetated wetlands F5 and F6 showed significant ($p < 0.05$) drops in turbidity at seven days of treatment. For LC–SGW, outflow means of TSS and turbidity decreased significantly for wetlands F4, F7 and F8, except for the vegetated wetlands F3, which showed no significant ($p > 0.05$) drop in TSS and turbidity after two days of treatment (Table 5.6).

After seven days of treatment for HC–SGW, the TSS and turbidity values dropped significantly ($p < 0.05$) at the presence of vegetation for wetlands F5 compared with those values for unvegetated wetlands F6. There was no significant ($p > 0.05$) effect of vegetation for the treatment of LC–SGW (Table 5.7). Increasing treatment time affected significantly ($p < 0.05$) decreasing TSS concentrations in the outflows of unvegetated wetlands F6 treating HC–SGW compared to those concentrations in wetlands F2 (2–day contact time). However, no significant ($p > 005$) effect of treatment time in terms of

turbidity was calculated for both systems. The TSS and turbidity values decreased significantly ($p < 0.05$) with increasing treatment time in the outflows of LC–SGW treated by both vegetated and unvegetated wetlands compared to wetlands operating at 2–day contact time (Table 5.7).

In general, the outflow mean values obtained for both TSS and turbidity ranked in this order: F5 < F1 < F6 < F2 (for wetlands treating HC–SGW) and F8 < F7 < F4 < F3 (for wetlands treated LC–SGW) in this order (Tables 5.3 and 5.4). For control wetlands containing tap water, the presence of macrophytes in C1 and C3 affected significantly ($p < 0.05$) both TSS and turbidity for both contact times compared to those values of unvegetated wetlands C2 and C4, respectively. However, an increase of contact time for the control wetland had no impact on TSS and turbidity (Table 5.7).

A comparison of the outflows of wetlands treating HC–SGW and LC–SGW revealed that there was no significant effect of the contaminant load strength on both TSS and turbidity removals from outflows of vegetated wetlands at both times of treatment. For unvegetated wetlands F2 and F6, removals of TSS and turbidity when treating HC–SGW at both contact times were significantly ($p < 0.05$) lower than those removals for corresponding wetlands treating LC–SGW F4 and F8, respectively (Table 5.8). In general, mean TSS and turbidity removals in FTW followed this order: F8 > F7 > F4 > F5 > F6 > F1 > F2 > F3, and F8 > F5 > F7 > F4 > F1 > F6 > F2 > F3, respectively (Tables 5.3 and 5.4).The correlation analysis results showed that TSS was significantly ($p < 0.01$) positively correlated with turbidity for all vegetated floating treatment wetland systems. The *P. australis* rhizome system and its attached biofilm enhances the processes of filtration and biodegradation supporting the removal of TSS and turbidity (Cheng et al., 2009).

5.3.4. The pH and Redox Potential

The pH is an important water quality parameter for aquatic treatment systems concerning both biological and chemical processes. During this study, the pH had a mean value of 8.4±1.61 for inflow HC–SGW, while it was close to neutral (6.9±0.48) for the inflow LC–SGW. The outflow mean values for all FTW (F1, F2, F5 and F6) treating HC–SGW were significantly ($p < 0.05$) lower than the inflow mean value. However, only the unvegetated wetlands (F4 and F8) treating LC–SGW have shown significant ($p < 0.05$) increases in pH compared to those value of the inflow (Table 5.6).

The presence of vegetation in the floating wetlands F1 and F5 (treating HC–SGW for two and seven days, respectively) did not affect significantly the pH values compared to those values of the unvegetated wetlands F2 (2–day contact time) and F6 (7–day contact time), respectively. In contrast, pH values decreased significantly ($p < 0.05$) at the presence of vegetation in wetlands treating LC–SGW F3 and F7 at both contact times compared to those outflow values for unvegetated wetlands F4 and F8, respectively (Table 5.7). The lower values of pH in LC–SGW effluents were fully compliant with those indicated by Ijaz et al. (2015). This can be explained by the release of carbon dioxide and acidic exudates produced from respiration of rhizomes. Furthermore, organic degradation by microorganisms can decrease pH through the generation of organic acids (Bezbaruah and Zhang, 2004; Sooknah and Wilkie, 2004; Iamchaturapatr et al., 2007; Lynch et al., 2015).

Regarding to the impact of increasing treatment contact time, the pH values of wetlands at 7–day contact time (F5, F6, F7 and F8) have not significantly changed compared to those outflow values for wetlands at 2–day contact time (F1, F2, F3 and F4, respectively) as indicated in Table 5.7. Furthermore, the highest pH values were recorded

at the end of the growth season for *P. australis* treating HC–SGW in wetlands F1 (2–day contact time) and F5 (7–day contact time). There are many possible reasons for this observation such as the release of gases during the degradation of dead plant tissues into the water column.

Temperatures below 4°C lead to an indirect increase of oxygen availability, which is not anymore required for microorganism activities. In addition, metabolism activities of aquatic organisms, macrophyte photosynthetic activities, consumption of nutrients and carbonate components can lead to an increase of pH due to the production of carbon dioxide for cell growth, and hydroxyl ions are left in excess (Wallace et al., 2016). In addition, a weakly negative correlation between COD and pH may be observed suggesting that the removal of organic matter through settling and biological conversion is likely to be associated with a rise in pH (Hamersley and Howes, 2002). Furthermore, agitation of the outflow during sampling contributes to the re–suspension of some salt particles.

For control wetlands containing tap water, the mean pH values decreased significantly (<0.05) at the presence of macrophytes for both contact times regarding wetlands C1 and C3 compared with the unvegetated wetlands C2 and C4, respectively. Only the unvegetated wetlands C4 have shown significant ($p < 0.05$) decreases in pH values with increasing contact time compared to those values of unvegetated wetlands C2 at 2–day contact time of treatment (Table 5.7). This is because of the significantly negative correlation relationship of both inflow and outflow pH with Eh values for all floating treatment wetland systems. So, outflow Eh values follow an inverse pattern concerning correlation relationships with pH values (Tables 5.6 and 5.7).

5.3.5. Dissolved Oxygen

The statistical analysis results indicated significant ($p < 0.05$) drops in the mean outflow DO concentrations at the presence of vegetation for wetlands treating HC–SGW (F1 and F5) and LC–SGW (F3 and F7) at both contact times of treatment compared to those concentrations of inflow HC–SGW and LC–SGW, respectively (Table 5.6). However, the DO was stable for wetland systems of the same design parameters, except for those systems without *P. australis* (F2 and F4). In other words, the presence of vegetation in wetlands affected significantly ($p < 0.05$) decreasing DO concentrations in the outflow of both types of greywater (F1, F3, F5 and F7) and at both contact times compared to those concentrations in the outflow of unvegetated wetlands (F2, F4, F6 and F8, respectively) as indicated in Table 5.7. These findings confirmed what previous researchers reported (Sooknah and Wilkie, 2004; Iamchaturapatr et al., 2007). However, Tanner et al. (1995) claimed that the existence of macrophytes in wetlands has a slight effect on outflow DO concentrations (Tanner, et al., 1995).

An increase in contact time affected significantly increasing DO concentrations in the outflows of unvegetated wetlands (F6 and F8) treating both types of greywater compared to those concentrations in the outflow of wetlands F2 and F4 (2–day contact time), respectively, which had the same design parameters (Table 5.7). The observed increase in DO with increasing contact time of treatment has been confirmed in previous studies by Tanner et al. (1995).

For control wetlands (Table 5.5), the mean DO concentrations of the outflow showed that the presence of macrophytes in wetlands C1 and C3 significantly ($p < 0.05$) impacted on the decreasing DO concentrations at both times of treatment compared to those concentrations of unvegetated wetlands C2 and C4, respectively (Table 5.6).

However, increasing contact times significantly ($p < 0.05$) influenced increasing DO concentrations in the outflow of unvegetated wetlands C4 compared to those concentrations of wetlands C2 with 2–day contact time (Table 5.7). This can be explained by microorganisms consuming oxygen for metabolism and degradation activities on biofilms located on surfaces provided by *P. australis* rhizome systems (Gersberg et al., 1986; Scholz et al., 2007; Van de Moortel, et al., 2010; Vymazal, 2011).

The statistical analysis for the effect of contamination load strengths showed that consumption rates of DO from the outflow of vegetated (F5) and unvegetated (F6) wetlands treating HC–SGW for 7–day contact time of treatment were significantly ($p < 0.05$) higher than those removals of wetlands treated LC–SGW F7 and F8, respectively (Table 5.8).

In case of significantly ($p < 0.05$) positive correlations of DO with both BOD_5 and COD, the sufficient supply of oxygen to aerobic heterotrophic bacteria will greatly increase the performance of aerobic biochemical oxidation. However, significantly ($p < 0.05$) negative correlations between DO and BOD_5 were observed for wetlands with no limited oxygen supply, where aerobic degradation is governed by the amount of active organic matter available to the organisms (Vymazal, 2005; 2010).

For most types of greywater, industrial wastewaters and storm water runoffs, DO correlates negatively with COD, because the supply of dissolved organic matter is not sufficient and aerobic degradation is not limited by DO (X. Li et al., 2009; Vymazal and Kröpfelová, 2009).

5.4. Growth Rate of *P. australis*

The growth of macrophytes in floating wetlands of both greywater types showed yellowish leaves during the experiment at low temperatures. The purification processes of traditional ecological floating wetlands have numerous disadvantages, including low purification efficiency at low temperature, and limited stand biomass and growth rates of the plants used (Billore et al., 2007; Faulwetter et al., 2011; Cao et al., 2016). Increasing DO values in the treatment system (Tables 5.3–5.5) were due to low water temperature in addition to the role of photosynthetic activity by macrophytes.

The DO significantly ($p < 0.05$) negatively correlated with temperature as confirmed by other researches (Cao et al., 2016). Positive correlations between pH and DO were observed at almost all FTW over the experimental period. However, Zhang et al. (2011) found no correlations between pH and DO. A high pH has been shown to inhibit photosynthesis causing a reduction in the macrophyte growth rate coupled with low temperature conditions (C. Zhang et al., 2014). Considerably lower growth rates were observed for the hydroponic plant rhizomes of *P. australis*, which might be an effect of light–induced fluorescein degradation due to partly open water surfaces (Seeger et al., 2013). Furthermore, the rhizomes of macrophytes are suspended in the water, and can therefore easily be damaged, consequently affecting negatively the removal performances in treatment systems.

Chapter Six

Treatment of Synthetic Greywater Using Pelletised Minewater Sludge (Ochre)

Abed, S.N., Almuktar, S.A, Scholz, M., 2017. Treatment of contaminated grey water using pelletised mine water sludge. Journal of Environmental Management, 197, 10–23.

6.1. Overview

In this chapter, mesocosm systems contained only synthetic greywater and those treated the synthetic greywaters with only cement–ochre pellets (T3, T4, T7, T8, T11, T12, T15 and T16) were considered as a simulation of stabilisation ponds and mentioned as (P1, P2, P3, P4, P5, P6, P7 and P8, respectively), as shown in Table 6.1.

Table 6.1. Operational variables in the experimental set–up designs of various mesocosm–scale constructed stabilisation ponds (P).

Pond[a]	Original label	Contact time		Water type		Cement–ochre pellets	
		Two days	*Seven days*	*High concentration*	*Low concentration*	*With*	*Without*
P1	T3	✓		✓			✓
P2	T4	✓		✓		✓	
P3	T7	✓			✓		✓
P4	T8	✓			✓	✓	
P5	T11		✓	✓			✓
P6	T12		✓	✓		✓	
P7	T15		✓		✓		✓
P8	T16		✓		✓	✓	

[a] Ponds P1 to P8 have four replicates each.

The inflow water quality was compared with the quality of outflow from each former systems to study the performance of each one. The effect of experimental variables such as presence of ochre pellets, contact time and concentrations of pollutant were investigated by comparison of outflows from treatment systems of ochre pellets, P2 (T4), P4 (T8), P6 (T12) and P8 (T16) with each other, from one side, and with outflows

of corresponding treatment system which contained only greywater, P1 (T3), P3 (T7), P5 (T11) and P7 (T15), respectively, discussed in chapter four.

To cover objective 3 of this study, section 1.5, characteristics of ochre sludge generated from Deerplay minewater treatment plant, considered in this study, was discussed and compared with other studies. The possibility of recycle ochre sludge or pellets made off was also investigated and compared with published literatures, in terms of release toxic trace elements into water, when it used for aqueous treatment systems. The remediation of synthetic greywater, phosphate and trace elements in particularly, and concentrations of adsorb trace elements which accumulated into cement–ochre pellets were also included in this chapter.

6.2. Ochre Characterisation

Ochre is brown or yellowish–brown indicating the presence of goethite (FeO(OH)) and smaller amounts of sulphur as reported by Singh et al. (1999), who proved that the ochre colour is correlated with the presence of certain minerals. Ochre consists of a mixture of ferrous components, but goethite is the most stable compound linked with acid mine drainage (Heal et al., 2005). The collected ochre had a moisture content of around 86% with pH and Eh values ranging between 6.4–7.1 and 27.2–32.5 mV, respectively.

The electrical conductivity of the tested ochre was between 323 and 481 µS/cm. These results indicate that the pH of acid mine water has been adjusted chemically by adding reagents through the treatment process and/or naturally during the oxidation process through treatment of mine water in constructed wetlands as practices at the Deerplay site (Barnes, 2000). Ferric iron precipitates out of water, which results in the formation of an iron precipitate and accumulation in the form of ochre.

According to Heal et al. (2003), chemical and physical characteristics of ochre vary with time as well with site location. A variety of potential recycling purposes have been proposed for ochre ranging from use as a cement pigment, a raw material for the steel industry, a soil amendment and a reagent for wastewater treatment. Since treating mine water results in an iron–rich sludge associated with Al, magnesium (Mg) and calcium (Ca), the option to exploit it for remediation of wastewater, particularly in phosphorus removal, is strongly recommended (Heal et al., 2005). Since fine particles of dried ochre might rapidly clog and harm treatment in this study, Portland cement was used to create granular solids from ochre sludge (section 3.3).

These cement–ochre solids have pH values between 9.63 and 12.53, and associated redox potentials between -248.5 and -98.8 mV. The average electrical conductivity range was 1580–2300 µS/cm.

The elemental analysis of digested samples for oven–dried raw ochre showed that ochre has a majority proportion of 47.0% iron (Fe), followed by Ca, Mg and aluminium (Al) of 2.6, 0.4 and 0.2%, respectively (Table 6.2). In comparison with findings published by Heal et al. (2005), The Fe, Ca, Mg and Al concentrations of Deerplay ochre were much lower than the corresponding concentrations in ochre obtained from Polkemmet (65.0, 7.0, 0.6 and 0.7%, respectively) and Minto sites (67.5, 11.8, 0.8 and 0.1% in that order).

Furthermore, the tested trace element concentrations for Deerplay ochre were less than the minimum concentration limits listed in USEPA Part 503–Tables I and III for biosolids, which regulate the disposal of sewage and industrial sludge solids in contact with the natural environment (USEPA, 1995).

Furthermore, considering Figure 6.1–a, Deerplay ochre sludge was compared with those ochres originated from other mine water treatment plants in the UK in terms of trace elements (Sapsford et al., 2015). The analytical results of Deerplay sludge ochre have shown that the Fe concentration was approximately equal to those concentrations of ochres from Glyncastle, Ynysarwed and Taff Merthyr mines. However, it was higher in Fe compared to other sites. Also, the Ca content was higher than that from ochres obtained from Taff Merthyr, Lindsay and Ynysarwed. The Mg and Al concentrations were lower than those from all other sites.

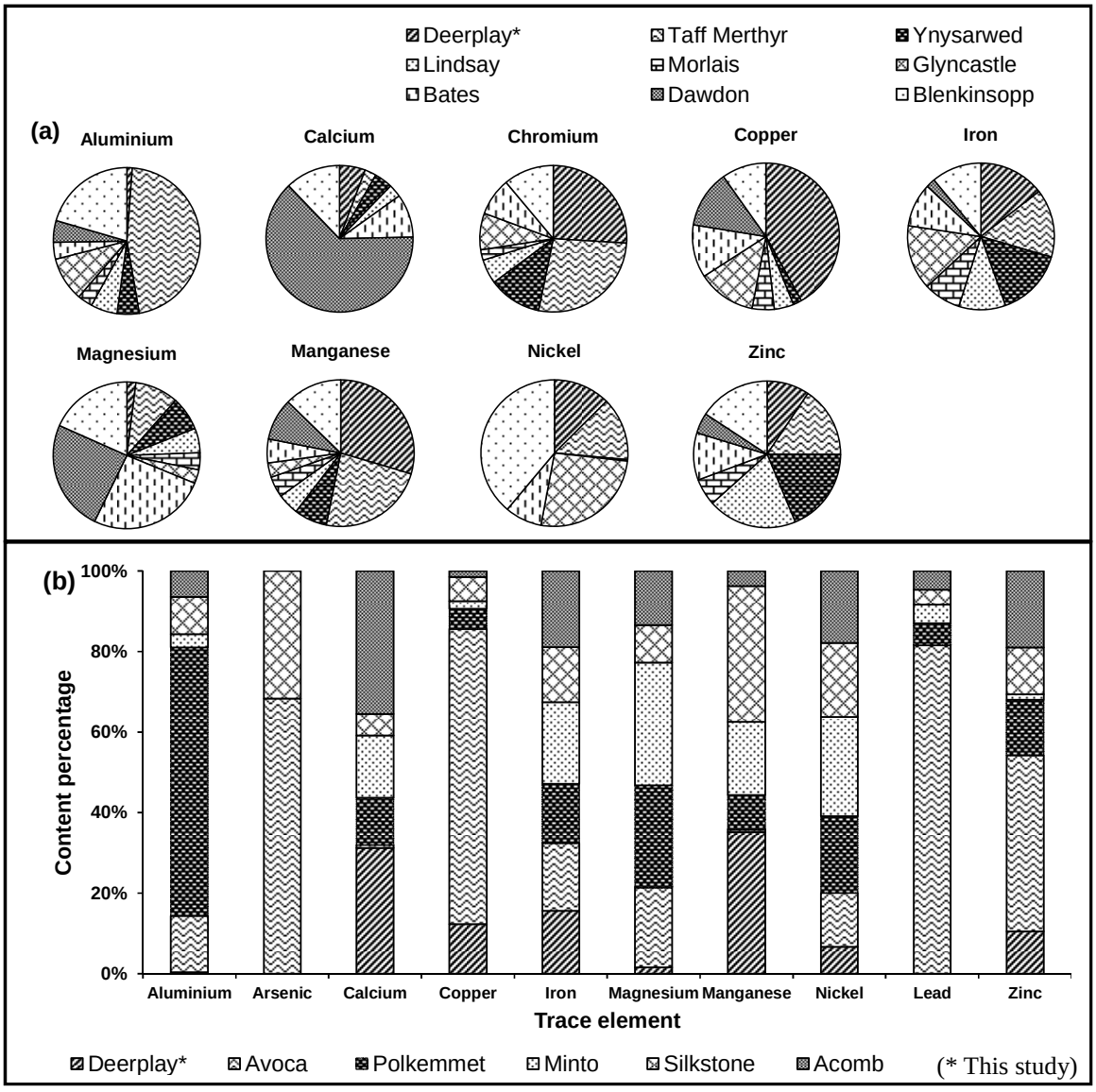


Figure 6.1. Comparison between the raw ochre sludge obtained from the Deerplay minewater treatment site and other sites in the United Kingdom considered by (a) Sapsford et al. (2015) and (b) Carr (2012).

In contrast, Cu and Mn concentrations in Deerplay ochre were highest. In addition, the Ni content was approximately equal to Taff Merthyr, but higher than those concentrations in ochres from Ynysarwed and Bates, and lower than those obtained from Glyncastle and Blenkinsopp mines. The Cr concentration in Deerplay sludge was lower than that in sludge from Taff Merthyr. The Zn content was higher compared to those in Dawdon and Morlais mines.

In Figure 6.1–b, another comparison of the trace element contents in Deerplay ochre with those of ochres from other mine sites in different UK locations were investigated by Carr (2012). The concentration of Fe in Deerplay ochre was lower than those in ochres from Avoca, Acomb and Minto, but greater than the Fe content from all other sites.

The Ca concentrations in ochres obtained from all sites (except for Acomb) were lower than that of Deerplay ochre. The highest Cu concentration was found for ochres of Avoca, but Deerplay ochre had greater Cu content compared to other sites. Minto ochres contained higher Zn concentrations, but those levels in ochre from other sites were less than that in Deerplay ochre. Furthermore, the Mn content percentage in Deerplay ochre was higher than that for all other sites, but approximately equal to that found in Silkstone's ochres. In contract, Al, Mg and Ni concentrations in Deerplay sludge were lower than those in ochres for all other sites. The concentrations of As and Pb regarding Deerplay ochre were rather low and below the detection limits of 2.0 mg/kg (USEPA, 1994). The analysis of digested samples of cement–ochre pellets indicated that the concentrations of Al, Ca, Cr, Mg and Zn were higher than those in raw sludge ochre, because they are normally included in Portland cement. However, concentrations of Cd, Fe and Mn reduced by approximately half compared to those concentrations found in ochre sludge (Table 6.2 and Figure 6.2), since the cement and ochre ratios in the mixture

were approximately equal (section 3.3.2). There were no changes in concentrations of B and Ni in cement–ochre pellet samples compared with those concentrations of raw ochre sludge samples.

Table 6.2. Trace element concentrations (mg/kg) detected in digested samples of raw ochre sludge and pellets produced from adding Portland cement to ochre sludge.

Element	Raw sludge ochre					Cement–ochre pellets				
	n	Mean	SD	Min	Max	n	Mean	SD	Min	Max
Aluminium	18	177.8	124.74	<0.001	345.7	18	13696.4	686.05	12448.4	14941.7
Boron	15	37.0	37.71	5.9	109.3	15	36.9	16.22	17.5	62.8
Calcium	21	25677.4	2733.47	19218.7	30147.8	21	268527.3	11891.67	245019.6	290052.8
Cadmium	16	8.7	11.41	<0.001	33.9	18	4.5	6.31	<0.001	18.9
Chromium	27	22.5	32.0	<0.001	112.0	27	47.5	24.99	17.9	109.1
Copper	27	95.7	127.98	<0.001	439.1	27	74.8	59.27	26.3	216.2
Iron	24	470458.5	79408.35	336308.0	578228.0	24	224603.5	10102.35	204262.0	240776.8
Magnesium	27	286.6	96.66	123.7	543.2	27	4859.0	338.98	3932.9	5281.4
Manganese	27	4276.3	720.18	3184.7	5430.5	27	2171.8	131.56	1951.6	2492.9
Nickel	16	15.8	10.48	2.3	34.4	17	15.5	10.44	0.7	32.8
Zinc	24	70.4	86.29	<0.001	394.0	24	447.7	149.16	216.3	783.1

Note: n, number of tested samples; SD, standard deviation; Min, minimum; and Max, maximum.

According to Hancock (2005), Cd concentrations in ochres from the UK are below 10 mg/kg. A relatively high concentration of nickel (1545 mg/kg) was found in Mouse Water ochre. Furthermore, Mn concentrations were relatively high in Polkemmet ochre. Deerplay ochre has been classified as non–hazardous concerning landfill (USEPA, 1995) and surface water regulations (FAO, 2003).

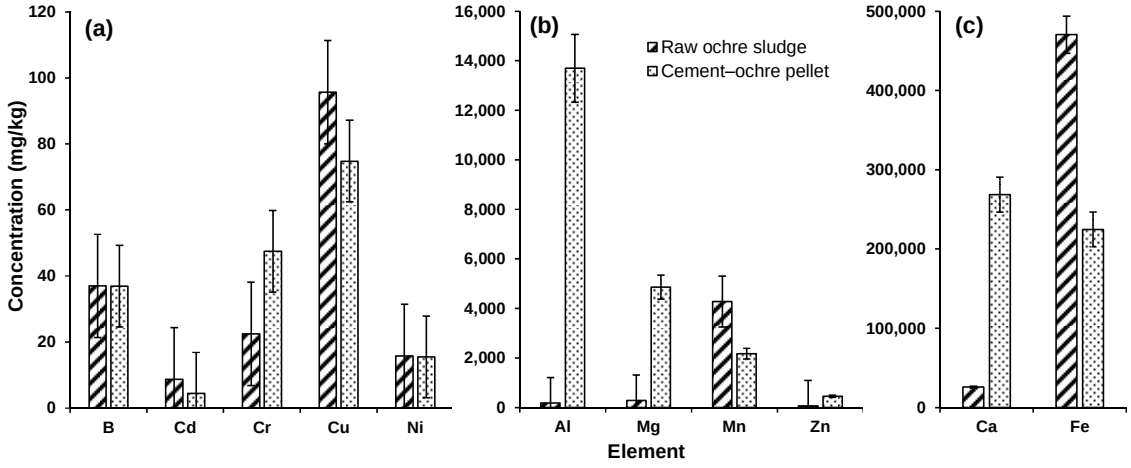


Figure 6.2. Comparison between the trace element content of the raw ochre sludge and pellets produced by adding Portland cement to ochre sludge before usage in the treatment of synthetic greywater.

Toxic metals may be released from abandoned mine sites into water or adsorbed onto iron within the settled ochre sludge at mine water treatment plants. There are concerns that toxic elements may mobilise from ochre–based materials into the aquatic environment (Carr, 2012). Ochre sludge, which is classified as non–hazardous, can be used in water remediation systems (Hancock, 2005). Furthermore, treatment processes must not impair the quality of treated water, which is designated for reuse or disposal to the environment (Sibrell et al., 2009).

In order to investigate contamination by ochre, dried samples of raw ochre sludge and cement–ochre pellets were brought individually into contact with deionised water. This was followed be the elemental analysis of that water for metal content (Dobbie et al., 2009).

The analysis of water samples was carried out after contact experiments shown in Table 6.3. Contact conditions simulated the worst case scenario by intensively shaking of proportional weights of solids and deionised water compared with typical water treatment conditions (section 3.3.3).

Table 6.3. Trace element concentrations (mg/l per kg of dry solids) released into deionised water from solids.

Element	Water samples with									
	Raw sludge ochre					*Cement–ochre solids*				
	n	Mean	SD	Min	Max	n	Mean	SD	Min	Max
Aluminium	–	nd	–	–	–	10	0.16	0.076	0.12	0.30
Boron	10	0.02	0.006	0.01	0.03	10	0.01	0.005	<0.001	0.02
Calcium	10	57.59	6.155	45.83	64.13	10	474.10	164.870	163.51	585.54
Cadmium	12	<0.001	0.005	<0.001	0.02	–	nd	–	–	–
Chromium	16	<0.001	0.002	<0.001	0.01	16	0.01	0.006	<0.001	0.02
Copper	–	nd	–	–	–	18	0.01	0.007	<0.001	0.02
Iron	14	0.97	0.173	0.78	1.15	14	0.19	0.005	0.18	0.20
Potassium	4	1.69	0.251	1.41	2.00	4	263.63	43.817	225.18	314.31
Magnesium	12	0.09	0.017	0.05	0.11	12	0.01	0.012	<0.001	0.04
Manganese	18	<0.001	0.007	<0.001	0.02	18	0.01	0.014	<0.001	0.04
Sodium	4	3.75	0.220	3.49	4.02	4	32.22	0.487	31.66	32.84
Nickel	–	nd	–	–	–	14	<0.001	0.001	<0.001	0.01
Zinc	–	nd	–	–	–	10	0.01	0.008	<0.001	0.02

Note: n, number of tested samples; SD, standard deviation; Min, minimum; Max, maximum; and nd, not detected when concentration < 0.001 mg/l.

The concentrations of released elements from both ochre and cement pellets were compared with limitations published by SEPA (2002) and FAO (2003), which concern the disposal of wastewater to surface water bodies and the reuse of wastewater for long–term irrigation, respectively. The concentrations of all released elements for both solids were below the thresholds set by these guidance documents, except for Al, which was released from cement–ochre pellets at a concentration of 0.16 mg/l, which was greater than the concentration limit set for surface water (0.025 mg/l) according to SEPA (2002). However, it was much lower than the requirements of irrigation water (5.0 mg/l) as set by FAO (2003).

The released Fe and Mg concentrations from ochre pellets were higher than those from raw ochre. However, both of them were less than the threshold set for surface water (2.0 mg/l) and irrigation purposes (5.0 mg/l) according to SEPA (2002) and FAO (2003), respectively. The released concentrations of Al, Cd, Cr, Cu, Mn, Ni and Zn from raw ochre were below the detection limits of the instrument, which were 45.0, 3.4, 6.1, 5.4, 1.4, 15.0 and 1.8 µg/l, respectively. Furthermore, Cd and Ni were below the detection limits of 3.4 and 15.0 µg/l, respectively, for water in contact with cement–ochre pellets (USEPA, 1994). The results show that there are no negative impacts on water quality, if ochre sludge from coal mine drainage and its corresponding cement pellets are used for water treatment purposes. This is in contrast to other waste solids such as steel slag and fly ash, which contain varying concentrations of contaminations such as Cd and As (Carr, 2012). In general, ochres generated from coal mining works have lower concentrations of toxic elements compared with ochres originating from metal mines (Sibrell et al., 2009). So, ochres, which are classified as non–hazardous, can be considered for application in wastewater treatment systems (Heal et al., 2005; Carr, 2012).

6.3. Inflow Greywater Quality

Table 6.4 shows the overall inflow water quality of both LC–SGW and HC–SGW for different loads of pollutants, which discussed in chapter four. Various outflow parameters were compared to assess potentially significant differences to highlight the effect of experimental operational parameters such as the presence of cement–ochre pellets, increase in contact time of treatment and load strength of pollutants in greywater.

Table 6.4. Physiochemical characteristics of inflow synthetic greywater (SGW).

Parameter	Unit	Inflow (low concentration)					Inflow (high concentration)				
		n	Mean	SD	Min	Max	n	Mean	SD	Min	Max
pH	–	81	6.9	0.48	5.3	7.9	81	8.4	1.61	5.4	11.5
Redox potential	mV	81	34.1	21.23	-18.1	111.2	81	-36.6	74.22	-182.1	97.9
Turbidity	NTU	81	22.9	7.14	9.8	41.6	81	188.9	47.22	18.3	308.0
Total suspended solids	mg/l	81	39.9	15.94	10.0	87.0	81	317.0	58.35	173.0	473.0
Electrical conductivity	µS/cm	81	164.6	63.24	98.7	452.0	81	988.5	196.09	612.0	1677.0
Dissolved oxygen	mg/l	81	10.4	1.24	7.7	12.3	81	10.5	1.39	6.9	12.6
Colour	Pa/Co	81	214.5	64.07	26.0	340.0	81	1587.8	379.89	787.0	2499.0
Temperature	°C	81	17.7	4.58	6.7	27.0	81	16.9	5.40	6.5	27.8
Biochemical oxygen demand	mg/l	81	17.6	8.00	2.0	40.0	81	34.7	12.99	10.0	60.0
Chemical oxygen demand	mg/l	81	28.9	14.47	8.2	86.7	81	129.2	34.68	63.9	221.0
Ammonia–nitrogen	mg/l	81	0.2	0.22	0.0	1.1	81	0.4	0.19	0.1	1.1
Nitrate–nitrogen	mg/l	81	1.3	1.21	0.1	7.6	81	8.9	6.38	0.2	29.8
Ortho–phosphate–phosphorus	mg/l	81	8.4	4.36	3.3	27.4	81	59.1	14.16	30.6	94.2
Element											
Aluminium	mg/l	45	0.52	0.528	0.09	1.56	45	2.13	0.869	0.76	4.77
Boron	mg/l	33	0.14	0.067	0.10	0.36	33	0.57	0.068	0.49	0.72
Calcium	mg/l	55	10.54	0.853	8.51	11.81	45	36.08	8.750	22.38	50.72
Cadmium	mg/l	42	0.09	0.056	0.04	0.23	39	7.36	2.981	3.67	11.58
Chromium	mg/l	54	0.04	0.063	0.01	0.23	54	3.20	0.918	1.67	5.70
Copper	mg/l	63	0.16	0.058	0.10	0.30	63	1.44	0.435	0.95	2.03
Iron	mg/l	48	0.21	0.102	0.11	0.43	48	6.41	2.476	2.48	10.37
Potassium	mg/l	12	4.04	0.448	3.41	4.65	12	60.16	1.684	58.31	62.53
Magnesium	mg/l	48	1.45	0.191	1.19	1.78	48	17.16	2.119	12.68	20.33
Manganese	mg/l	63	0.17	0.084	0.07	0.26	63	0.98	0.257	0.64	1.29
Sodium	mg/l	12	14.32	1.662	12.01	16.00	12	62.68	14.538	45.50	76.92
Nickel	mg/l	51	0.04	0.065	0.01	0.18	51	0.05	0.065	0.01	0.20
Zinc	mg/l	39	0.21	0.159	0.01	0.39	39	4.25	1.500	2.22	6.34

Note: n, number of tested samples; SD, standard deviation; Min, minimum; Max, maximum; and NTU, nephelometric turbidity unit.

6.4. Outflow Greywater Quality

6.4.1. Removal of Ortho–Phosphate–Phosphorus (PO_4–P)

Test results of the outflow greywater quality are shown in Table 6.5. In terms of PO_4–P, the statistical analysis showed that the outflows from all ponds (except for P7 treating LC–SGW without ochre pellets) were significantly lower than the corresponding SGW inflow concentrations (Table 6.6).

The PO_4–P removals in systems without ochre pellets can be explained by either sedimentation and/or microorganism activities. However, there are not sufficient nutrients in LC–SGW for the survival of organisms, which are limited by PO_4–P removal (Jefferson et al., 2001). This scenario could be the case for pond P7.

If cement–ochre pellets are present in treatment systems, both types of SGW showed significant removal rates in PO_4–P compared with those removals in ponds without ochre pellets (Table 6.7). In addition, the values of pH, Eh and EC significantly ($p < 0.05$) changed, if ochre pellets are present in ponds treating both types of SGW (P2, P4, P6 and P8) and at both contact times compared with ponds without cement–ochre pellets (P1, P3, P5 and P7, respectively).

However, no significant ($p > 0.05$) differences in outflow EC values concerning ponds treating HC–SGW at 7–day contact time for P5 (without ochre pellets) compared to P6 (with ochre pellets) were noted. However, an increase in contact time for HC–SGW treatment has significantly affected the change in pH and Eh values in the presence of ochre pellets for the outflow of P6 compared with the outflow of P2 (2–day contact time with ochre pellets), as shown in Table 6.7.

Because the cement–ochre pellets are associated with a relatively high pH value, the corresponding pH of the greywater increased significantly ($p < 0.05$) at the presence of these pellets during treatment. The PO_4–P removals also increased significantly ($p < 0.05$) for these treatments. A negative correlation between pH and PO_4–P ($r = -0.265$, $p < 0.001$) has been recorded.

Reductions in PO_4–P can be explained either by adsorption processes linked to ochre pellets or precipitation processes due to the high concentration of Ca, which converts the dissolved PO_4–P to insoluble substances under alkaline conditions (Heal et al., 2003; Dobbie et al., 2009).

The treated SGW in this study was agitated before sampling to evaluate the pollutant removal mechanisms other than sedimentation, which included adsorption by ochre pellets. Therefore, calcium phosphate precipitation was the dominant removal process for the cement–ochre pellets with a minor proportion of removal associated to the ochre sludge (Littler et al., 2013).

The results revealed that the usage of cement–ochre pellets led to a significant ($p < 0.05$) increase in turbidity, TSS and colour for all ponds treating both types of SGW at both times of treatment (P2, P4, P6 and P8) compared to the outflows of ponds without ochre pellets (P1, P3, P5 and P7, respectively). An increase in contact time in the treatment of HC–SGW with ochre pellets results in a significant ($p < 0.05$) increase in turbidity and TSS.

However, significant ($p < 0.05$) decreases of these variables were recorded for ponds without ochre pellets, especially for ponds treating LC–SGW. This could be explained by the negative correlations between PO_4–P and both turbidity ($r = -0.403$, $p = 0.018$) and TSS ($r = -0.367$, $p = 0.033$).

Table 6.5. Outflow water quality parameters for 2–day and 7–day contact times in ponds (P) for both high (HC) and low (LC) contaminations of synthetic greywater (SGW).

Parameter	Unit	2–day outflow (HC)–P1						2–day outflow (HC)–P2					
		n	Mean	SD	Min	Max	Rem (%)	n	Mean	SD	Min	Max	Rem (%)
pH	–	85	7.8	1.37	5.6	9.8	na	36	8.7	1.73	6.6	12.7	na
Redox potential	mV	85	-3.0	62.95	-107.6	88.6	na	36	-49.9	83.61	-232.8	42.1	na
Turbidity	NTU	85	192.1	50.87	102.0	341.0	-1.7	36	191.3	84.41	106.0	456.0	-1.3
Total suspended solids	mg/l	85	321.8	56.68	165.0	447.0	-1.5	36	337.4	109.45	161.0	661.0	-6.4
Electrical conductivity	µS/cm	85	965.2	106.68	627.0	1208.0	na	36	1178.4	264.41	806.0	1944.0	na
Dissolved oxygen	mg/l	85	10.2	0.73	8.7	12.1	2.9	36	10.0	0.52	8.9	10.9	4.8
Colour	Pa/Co	85	1527.6	326.28	677.0	2311.0	3.8	36	1935.6	702.18	702.0	3438.0	-21.9
Temperature	°C	85	17.1	4.75	6.1	23.6	na	36	17.2	4.73	6.0	23.4	na
Biochemical oxygen demand	mg/l	85	14.7	7.78	0.0	40.0	57.6	36	11.7	7.71	0.0	35.0	66.3
Chemical oxygen demand	mg/l	85	106.6	22.68	43.3	164.0	17.5	36	100.3	21.08	41.7	131.0	22.4
Ammonia–nitrogen	mg/l	85	0.4	0.16	0.0	0.9	0.0	36	0.4	0.09	0.2	0.6	0.0
Nitrate–nitrogen	mg/l	85	9.4	4.67	0.5	24.0	-5.6	36	12.9	7.03	2.4	26.9	-44.9
Ortho–phosphate–phosphorus	mg/l	85	46.2	10.74	23.7	70.1	21.8	36	19.5	4.98	9.0	30.3	67.0
Element													
Aluminium	mg/l	39	2.41	1.016	0.74	4.25	-13.15	39	2.98	2.087	0.84	7.14	-39.91
Boron	mg/l	35	0.54	0.060	0.42	0.66	5.26	35	0.50	0.078	0.40	0.68	12.28
Calcium	mg/l	37	43.02	2.411	35.94	46.28	-19.24	37	104.13	32.868	46.28	141.65	-188.61
Cadmium	mg/l	42	7.69	1.064	4.95	8.98	-4.48	42	7.14	2.429	4.23	11.75	2.99
Chromium	mg/l	58	3.76	1.203	1.34	4.98	-17.5	58	3.99	1.806	1.87	7.03	-24.69
Copper	mg/l	63	1.45	0.113	1.28	1.70	-0.69	63	1.55	0.308	1.23	2.09	-7.64
Iron	mg/l	51	6.35	2.423	1.56	9.29	0.94	51	7.11	2.934	2.60	11.55	-10.92
Potassium	mg/l	14	55.68	4.486	49.48	60.69	7.45	14	60.47	15.561	49.85	85.46	-0.52
Magnesium	mg/l	48	17.76	1.392	13.92	19.55	-0.20	48	13.33	4.526	7.53	19.80	22.32
Manganese	mg/l	63	1.19	0.063	1.06	1.29	-21.4	63	0.89	0.396	0.61	1.59	9.18
Sodium	mg/l	14	58.19	10.620	42.35	68.22	7.16	14	58.54	11.630	41.37	76.33	6.60
Nickel	mg/l	53	0.03	0.018	0.00	0.06	40.00	53	0.03	0.033	0.00	0.10	40.00
Zinc	mg/l	44	4.30	0.524	3.12	5.25	-1.42	44	4.52	0.961	2.90	6.40	-6.35
		2–day outflow (LC)–P3						2–day outflow (LC)–P4					
pH	–	85	7.5	0.70	6.3	10.1	na	36	10.6	0.99	8.5	12.5	na
Redox potential	mV	85	4.2	30.40	-116.1	51.0	na	36	-143.5	51.01	-238.1	-30.3	na
Turbidity	NTU	85	20.2	14.20	2.9	129.0	11.8	36	35.6	18.11	8.7	79.3	-55.5
Total suspended solids	mg/l	85	30.0	12.12	11.0	76.0	24.8	36	66.2	36.63	13.0	181.0	-65.9
Electrical conductivity	µS/cm	85	138.5	23.26	79.0	215.0	na	36	344.5	287.03	168.4	1534.0	na
Dissolved oxygen	mg/l	85	10.5	0.82	8.2	12.6	-1.0	36	10.1	0.73	6.5	10.8	2.9
Colour	Pa/Co	85	164.5	40.93	34.0	265.0	23.3	36	331.7	119.34	104.0	552.0	-54.6
Temperature	°C	85	16.0	4.59	5.3	21.8	na	36	16.3	4.24	6.3	21.3	na
Biochemical oxygen demand	mg/l	85	5.6	3.60	0.0	20.0	68.2	36	4.4	5.13	0.0	22.0	75.0
Chemical oxygen demand	mg/l	85	26.8	6.18	15.4	41.9	7.3	36	24.0	4.99	15.4	39.9	17.0
Ammonia–nitrogen	mg/l	85	0.09	0.05	0.0	0.3	55.0	36	0.1	0.04	0.1	0.2	50.0
Nitrate–nitrogen	mg/l	85	1.2	0.71	0.1	3.2	7.7	36	0.6	0.54	0.0	2.6	53.8
Ortho–phosphate–phosphorus	mg/l	85	7.0	3.89	3.0	18.8	16.7	36	3.9	1.25	2.2	7.1	53.6
Element													
Aluminium	mg/l	39	0.34	0.180	0.11	0.72	34.62	39	0.76	0.347	0.16	1.24	-46.15
Boron	mg/l	35	0.11	0.009	0.08	0.13	21.43	35	0.10	0.024	0.07	0.13	28.57
Calcium	mg/l	46	11.25	0.773	9.86	12.70	-6.74	37	70.99	33.166	21.66	109.98	-573.53
Cadmium	mg/l	42	0.05	0.031	0.00	0.11	44.44	42	0.04	0.030	0.00	0.10	55.56
Chromium	mg/l	58	0.04	0.049	0.00	0.12	0.00	58	0.05	0.039	0.00	0.12	-25.00
Copper	mg/l	63	0.06	0.049	0.02	0.15	62.50	63	0.05	0.043	0.01	0.13	68.75
Iron	mg/l	51	0.21	0.157	0.09	0.45	0.00	51	0.48	0.447	0.15	1.26	-128.57
Potassium	mg/l	14	3.87	0.364	3.35	4.50	4.21	14	12.77	15.139	2.73	36.71	-216.09
Magnesium	mg/l	48	1.35	0.133	0.99	1.58	6.90	48	0.70	0.336	0.28	1.15	51.72
Manganese	mg/l	63	0.08	0.056	0.00	0.18	52.94	63	0.08	0.069	0.00	0.20	52.94
Sodium	mg/l	14	13.82	1.175	12.14	15.57	3.49	14	15.35	3.197	12.32	20.34	-7.19
Nickel	mg/l	53	0.01	0.007	0.00	0.04	75.00	53	0.01	0.012	0.00	0.06	75.00
Zinc	mg/l	42	0.09	0.083	0.00	0.23	57.14	42	0.07	0.084	0.00	0.29	66.67

Table 6.5. (Continued).

Parameter	Unit	7–day outflow (HC)–P5						7–day outflow (HC)–P6					
		n	Mean	SD	Min	Max	Rem (%)	n	Mean	SD	Min	Max	Rem (%)
pH	–	83	7.7	1.21	5.9	9.9	na	34	9.8	1.54	7.1	12.3	na
Redox potential	mV	83	-4.4	59.67	-108.3	78.1	na	34	-95.5	88.21	-216.7	157.8	na
Turbidity	NTU	83	185.7	49.24	65.1	281.0	1.7	34	245.8	96.29	60.0	497.0	-30.1
Total suspended solids	mg/l	83	302.6	61.44	147.0	434.0	4.5	34	423.4	114.04	120.0	692.0	-33.6
Electrical conductivity	µS/cm	83	1003.0	306.88	492.0	2460.0	na	34	1107.1	299.47	734.0	1814.0	na
Dissolved oxygen	mg/l	83	10.5	0.91	7.9	12.0	0.0	34	9.8	1.19	5.4	11.7	6.7
Colour	Pa/Co	83	1644.8	489.96	718.0	2889.0	-3.6	34	2040.5	757.57	688.0	3282.0	-28.5
Temperature	°C	83	16.6	3.87	7.6	22.1	na	34	17.7	4.20	7.4	22.3	na
Biochemical oxygen demand	mg/l	83	16.6	7.07	4.0	38.0	52.2	34	8.3	4.23	0.0	18.0	76.1
Chemical oxygen demand	mg/l	83	100.8	27.65	11.6	159.5	22.0	34	103.1	16.10	75.6	135.0	20.2
Ammonia–nitrogen	mg/l	83	0.3	0.13	0.0	0.8	25.0	34	0.3	0.11	0.1	0.5	25.0
Nitrate–nitrogen	mg/l	83	8.5	8.42	0.4	34.5	4.5	34	15.0	8.59	3.7	38.7	-68.5
Ortho–phosphate–phosphorus	mg/l	83	43.0	13.78	20.25	79.4	27.2	34	17.3	5.63	3.8	32.8	70.7
Element													
Aluminium	mg/l	54	2.98	1.218	1.61	6.14	-39.91	24	3.61	2.306	0.87	6.67	-69.48
Boron	mg/l	26	0.54	0.160	0.34	0.77	5.26	20	0.39	0.078	0.30	0.51	31.58
Calcium	mg/l	52	37.39	4.030	30.58	45.66	-3.63	22	145.67	92.506	40.36	243.66	-303.74
Cadmium	mg/l	36	6.40	1.984	3.86	9.72	13.59	24	6.87	2.628	3.33	10.27	6.66
Chromium	mg/l	46	4.76	1.215	2.83	6.68	-48.75	34	4.75	2.021	2.57	6.89	-48.44
Copper	mg/l	54	1.30	0.301	0.80	1.76	9.72	36	1.47	0.247	1.14	1.80	-2.08
Iron	mg/l	42	7.02	1.801	3.58	9.36	-9.52	30	8.69	2.012	6.48	10.99	-35.57
Potassium	mg/l	8	45.77	5.160	39.87	51.00	23.92	8	59.62	20.132	39.79	79.86	0.90
Magnesium	mg/l	54	16.24	1.971	11.76	18.35	5.36	30	12.97	3.785	7.71	17.80	24.42
Manganese	mg/l	54	1.01	0.223	0.75	1.38	-3.06	36	0.86	0.457	0.33	1.42	12.24
Sodium	mg/l	8	55.22	11.852	41.86	67.68	11.90	8	55.59	12.232	42.05	68.09	11.31
Nickel	mg/l	50	0.09	0.081	0.00	0.20	-80.00	32	0.04	0.033	0.00	0.10	20.00
Zinc	mg/l	32	3.90	0.972	1.90	5.10	8.02	26	4.32	0.787	3.01	5.69	-1.65
		7–day outflow (LC)– P7						7–day outflow (LC)–P8					
pH	–	83	7.5	0.72	6.4	9.3	na	34	10.5	1.05	8.0	12.2	na
Redox potential	mV	83	1.8	33.00	-87.9	53.2	na	34	-131.3	72.36	-217.6	156.0	na
Turbidity	NTU	83	16.5	7.27	5.7	34.1	27.9	34	40.9	25.03	4.0	113.0	-78.6
Total suspended solids	mg/l	83	25.0	10.96	7.0	56.0	37.3	34	55.2	24.85	4.0	104.0	-38.3
Electrical conductivity	µS/cm	83	144.0	32.28	97.7	263.0	na	34	290.2	135.74	148.0	768.0	na
Dissolved oxygen	mg/l	83	11.0	1.11	8.1	14.3	-5.8	34	10.1	0.84	8.3	11.2	2.9
Colour	Pa/Co	83	152.6	41.05	51.0	258.0	28.9	34	283.8	115.21	48.0	544.0	-32.3
Temperature	°C	83	15.3	4.23	6.7	22.2	na	34	17.0	4.15	7.9	22.1	na
Biochemical oxygen demand	mg/l	83	6.7	4.85	0.0	22.0	61.9	34	5.4	3.95	0.0	20.0	69.3
Chemical oxygen demand	mg/l	83	17.2	6.95	6.0	36.7	40.5	34	19.9	7.28	3.9	32.2	31.1
Ammonia–nitrogen	mg/l	83	0.1	0.04	0.0	0.3	50.0	34	0.1	0.15	0.0	0.8	50.0
Nitrate–nitrogen	mg/l	83	1.0	0.64	0.0	4.0	23.1	34	0.3	0.28	0.0	1.1	76.9
Ortho–phosphate–phosphorus	mg/l	83	8.5	4.03	2.6	19.6	-10.0	34	3.7	1.29	1.2	6.6	56.0
Element													
Aluminium	mg/l	44	0.36	0.189	0.09	0.75	30.77	24	0.73	0.420	0.20	1.40	-40.38
Boron	mg/l	24	0.12	0.064	0.08	0.26	14.29	20	0.08	0.006	0.07	0.09	42.86
Calcium	mg/l	52	10.74	0.739	9.44	12.12	-1.90	22	65.46	37.361	23.48	104.98	-521.06
Cadmium	mg/l	32	0.09	0.083	0.00	0.21	0.00	24	0.05	0.046	0.00	0.11	44.44
Chromium	mg/l	42	0.07	0.074	0.00	0.21	-75.00	34	0.06	0.054	0.00	0.12	-50.00
Copper	mg/l	48	0.10	0.091	0.00	0.26	37.50	36	0.06	0.057	0.00	0.13	62.50
Iron	mg/l	38	0.20	0.100	0.07	0.30	4.76	30	0.93	0.759	0.15	1.91	-342.86
Potassium	mg/l	8	3.62	0.438	3.07	4.22	10.40	8	20.16	19.003	2.26	38.75	-399.01
Magnesium	mg/l	46	1.38	0.161	1.03	1.64	4.83	30	0.78	0.330	0.36	1.16	46.21
Manganese	mg/l	48	0.06	0.074	0.00	0.21	64.71	36	0.10	0.094	0.00	0.21	41.18
Sodium	mg/l	8	13.15	1.199	11.83	14.36	8.17	8	15.69	5.272	10.55	21.03	-9.57
Nickel	mg/l	44	0.05	0.080	0.00	0.18	-25.00	32	0.01	0.010	0.00	0.05	75.00
Zinc	mg/l	28	0.13	0.068	0.01	0.25	38.10	24	0.11	0.089	0.00	0.26	47.62

Note: n, number of tested samples; SD, standard deviation; Min, minimum; Max, maximum; Rem, removal; NTU, nephelometric turbidity unit; and na, not applicable.

Table 6.6. Significant values of the statistical analysis concerning the comparison between treated and untreated greywater samples of; (a) high concentration synthetic greywater (HC–SGW), and (b) low concentration synthetic greywater (LC–SGW).

(a) HC–SGW (inflow and outflow)

		2–day contact time						**7–day contact time**					
		Inflow & P1			***Inflow & P2***			***Inflow & P5***			***Inflow & P6***		
Parameter	**Unit**	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
pH	–	<0.001	M–W	0.027	0.149	T–test	0.313	<0.001	M–W	0.004	0.021	M–W	<0.001
Redox potential	mV	<0.001	M–W	0.004	0.081	T–test	0.390	<0.001	M–W	0.004	0.099	T–test	<0.001
Turbidity	NTU	0.118	T–test	0.676	<0.001	M–W	0.071	0.471	T–test	0.667	<0.001	M–W	<0.001
Total suspended solids	mg/l	0.217	T–test	0.593	<0.001	M–W	0.603	0.318	T–test	0.127	<0.001	M–W	<0.001
Electrical conductivity	µS/cm	0.012	M–W	0.376	<0.001	M–W	<0.001	0.042	M–W	0.320	<0.001	M–W	0.149
Dissolved oxygen	mg/l	0.005	M–W	0.002	0.007	M–W	0.001	0.004	M–W	0.301	0.001	M–W	0.003
Colour	Pa/Co	0.218	T–test	0.274	<0.001	M–W	0.032	0.285	T–test	0.405	0.001	M–W	0.001
Temperature	°C	0.005	M–W	0.923	0.005	M–W	0.939	0.004	M–W	0.585	0.003	M–W	0.502
Biochemical oxygen demand	mg/l	0.002	M–W	<0.001	0.002	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Chemical oxygen demand	mg/l	0.005	M–W	<0.001	0.014	M–W	<0.001	0.002	M–W	<0.001	0.001	M–W	<0.001
Ammonia–nitrogen	mg/l	<0.001	M–W	0.860	<0.001	M–W	0.545	<0.001	M–W	0.091	<0.001	M–W	0.009
Nitrate–nitrogen	mg/l	0.005	M–W	0.325	<0.001	M–W	0.006	<0.001	M–W	0.204	<0.001	M–W	<0.001
Ortho–phosphate–phosphorus	mg/l	0.107	T–test	<0.001	<0.001	M–W	<0.001	0.195	T–test	<0.001	<0.001	M–W	<0.001
Aluminium	mg/l	0.009	M–W	0.171	<0.001	M–W	0.696	0.156	T–test	<0.001	<0.001	M–W	0.104
Boron	mg/l	0.002	M–W	0.171	0.004	M–W	<0.001	0.016	M–W	0.593	0.018	M–W	<0.001
Calcium	mg/l	0.001	M–W	0.001	<0.001	M–W	<0.001	0.003	M–W	0.068	<0.001	M–W	<0.001
Cadmium	mg/l	0.001	M–W	0.159	<0.001	M–W	0.128	<0.001	M–W	0.340	<0.001	M–W	0.119
Chromium	mg/l	0.004	M–W	0.003	<0.001	M–W	0.104	<0.001	M–W	<0.001	<0.001	M–W	0.119
Copper	mg/l	<0.001	M–W	0.378	<0.001	M–W	0.005	<0.001	M–W	0.177	<0.001	M–W	0.436
Iron	mg/l	0.002	M–W	0.867	<0.001	M–W	0.140	0.003	M–W	0.193	<0.001	M–W	<0.001
Potassium	mg/l	0.004	M–W	0.013	0.003	M–W	0.067	0.002	M–W	<0.001	0.011	M–W	1.000
Magnesium	mg/l	0.007	M–W	0.120	<0.001	M–W	<0.001	0.024	M–W	0.277	<0.001	M–W	<0.001
Manganese	mg/l	<0.001	M–W	0.006	<0.001	M–W	<0.001	<0.001	M–W	0.023	<0.001	M–W	0.595
Sodium	mg/l	0.007	M–W	0.322	0.009	M–W	0.432	0.005	M–W	0.181	0.004	M–W	0.181
Nickel	mg/l	<0.001	M–W	0.169	<0.001	M–W	0.077	<0.001	M–W	0.016	<0.001	M–W	0.574
Zinc	mg/l	0.005	M–W	0.500	<0.001	M–W	0.208	0.001	M–W	0.503	0.001	M–W	0.538

Note: p value, significant difference, if $p < 0.05$ and not significant if $p > 0.05$; M–W, Mann–Whitney U–test; and NTU, nephelometric turbidity unit.

Table 6.6. (Continued).

(b) LC–SGW (inflow and outflow)

Parameter	Unit	2–day contact time						7–day contact time					
		Inflow & P3			*Inflow & P4*			*Inflow & P7*			*Inflow & P8*		
		Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
pH	–	0.132	T–test	<0.001	<0.001	M–W	<0.001	0.005	M–W	<0.001	<0.001	M–W	<0.001
Redox potential	mV	0.013	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Turbidity	NTU	<0.001	M–W	0.004	<0.001	M–W	<0.001	0.008	M–W	<0.001	<0.001	M–W	<0.001
Total suspended solids	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	0.002	M–W	<0.001
Electrical conductivity	µS/cm	<0.001	M–W	0.001	<0.001	<0.001	M–W	<0.001	M–W	0.013	<0.001	M–W	<0.001
Dissolved oxygen	mg/l	<0.001	M–W	0.846	<0.001	M–W	0.009	<0.001	M–W	0.005	<0.001	M–W	0.068
Colour	Pa/Co	0.483	T–test	<0.001	0.003	M–W	<0.001	0.245	T–test	<0.001	0.100	T–test	0.002
Temperature	°C	0.015	M–W	0.023	0.009	M–W	0.096	0.002	M–W	0.001	0.002	M–W	0.392
Biochemical oxygen demand	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Chemical oxygen demand	mg/l	<0.001	M–W	0.811	<0.001	M–W	0.199	<0.001	M–W	<0.001	<0.001	M–W	0.001
Ammonia–nitrogen	mg/l	<0.001	M–W	<0.001	<0.001	M–W	0.037	<0.001	M–W	<0.001	<0.001	M–W	0.019
Nitrate–nitrogen	mg/l	<0.001	M–W	0.687	<0.001	M–W	<0.001	<0.001	M–W	0.201	<0.001	M–W	<0.001
Ortho–phosphate–phosphorus	mg/l	<0.001	M–W	0.003	<0.001	M–W	<0.001	<0.001	M–W	0.606	<0.001	M–W	<0.001
Aluminium	mg/l	<0.001	M–W	0.192	<0.001	M–W	0.080	<0.001	M–W	0.321	<0.001	M–W	0.025
Boron	mg/l	<0.001	M–W	0.001	<0.001	M–W	0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Calcium	mg/l	0.058	T–test	<0.001	<0.001	M–W	<0.001	0.003	M–W	0.295	<0.001	M–W	<0.001
Cadmium	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	0.290	<0.001	M–W	0.050
Chromium	mg/l	<0.001	M–W	0.014	<0.001	M–W	0.005	<0.001	M–W	0.360	<0.001	M–W	0.738
Copper	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Iron	mg/l	<0.001	M–W	0.001	<0.001	M–W	0.051	<0.001	M–W	0.444	<0.001	M–W	<0.001
Potassium	mg/l	0.125	T–test	0.309	0.536	T–test	0.050	0.334	T–test	0.054	<0.001	M–W	1.000
Magnesium	mg/l	<0.001	M–W	0.776	<0.001	M–W	0.041	<0.001	M–W	0.531	<0.001	M–W	0.041
Manganese	mg/l	<0.001	M–W	0.250	<0.001	M–W	0.031	<0.001	M–W	0.875	<0.001	M–W	0.031
Sodium	mg/l	0.010	M–W	0.403	0.043	M–W	0.705	0.012	M–W	0.181	0.028	M–W	1.000
Nickel	mg/l	<0.001	M–W	<0.001	<0.001	M–W	0.410	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Zinc	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	0.328	<0.001	M–W	0.010

Note: p value, significant difference, if $p < 0.05$ and not significant if $p > 0.05$; M–W, Mann–Whitney U–test; and NTU, nephelometric turbidity unit.

Oxides and/or hydroxides of Fe and Al released from ochre pellets, and compounds of Ca had the ability to adsorb phosphorus (Heal et al., 2005). Phosphorus concentrations can decrease within a few minutes in non–agitated conditions (McHaffie et al., 2001). Therefore, an increase in contact time does not necessarily lead to a decrease in PO_4–P, if ochre pellets are present. In terms of strengths of contamination load, PO_4–P was significantly ($p < 0.05$) removed at the presence of ochre pellets from HC–SGW compared with those removals from LC–SGW (Table 6.7 and Figure 6.3–a). In general, PO_4–P removal efficiencies in ponds ranked as follows: P6 > P2 > P8 > P4 > P5 > P1 > P3 > P7.

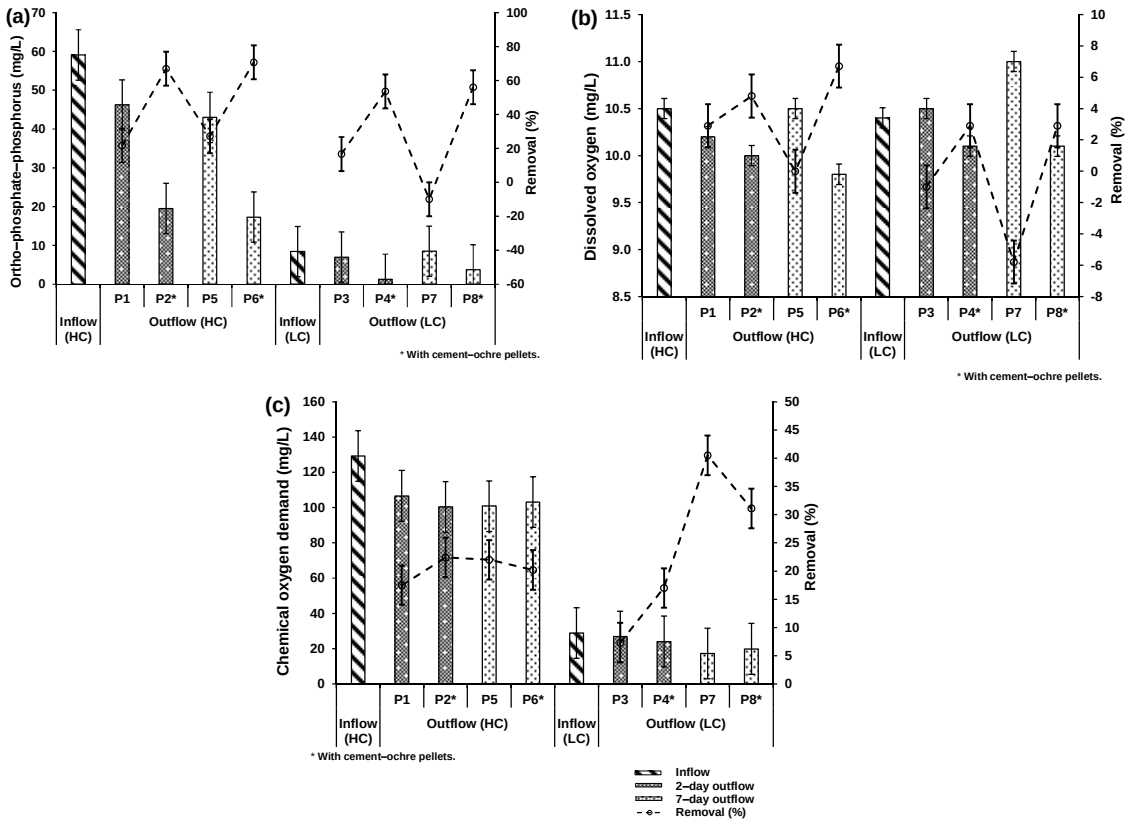


Figure 6.3. Comparison between inflow and outflow synthetic greywater treated in various experimental set–up designs of simulated stabilisation ponds (P) in terms of; (a) ortho–phosphate–phosphorus, (b) dissolved oxygen, and (c) chemical oxygen demand.

Littler et al. (2013) reported that cement–ochre pellets have the ability to continue to remove phosphorus in experiments of up to seven days, whereas the unbound ochre sludge approached an equilibrium in a phosphate solution after about 60 minutes of contact time.

Chemical oxidation processes involving elements including Fe, Al and Ca may consume dissolved oxygen in greywater, if cement–ochre pellets are present. Systems treating LC–SGW for two days with ochre pellets were associated with a significant ($p < 0.05$) decrease in DO and COD concentrations. In comparison, no significant ($p > 0.05$) effect on these parameters were recorded for the treatment of HC–SGW (Figures 6.3–b and c).

After seven days of contact time, the use of ochre pellets decreased the DO content significantly ($p < 0.05$) for HC–SGW with no effect on COD. However, there was a significant ($p < 0.05$) increase in COD and also a significant ($p < 0.05$) decrease in DO for the treatment of LC–SGW (Table 6.7). The content of PO_4–P negatively correlated with DO ($r = -0.226$, $p < 0.001$) and positively with COD ($r = 0.673$, $p < 0.001$).

Table 6.7. Significant values of the statistical analysis for treated greywater samples to investigate (a) the effects of the presence of cement–ochre pellets on changing values of parameters in ponds (P); (b) the effects of increasing contact time on changing values of parameters in ponds (P), and (c) the effects of pollutant loads on removal rates of contaminants in ponds (P).

(a) Effects of the presence of cement–ochre pellets on remediation in ponds (P)

		2–day contact time						**7–day contact time**					
		HC–SGW (P1 & P2)			***LC–SGW (P3 & P4)***			***HC–SGW (P5 & P6)***			***LC–SGW (P7 & P8)***		
Parameter	**Unit**	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
pH	–	<0.001	M–W	0.009	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Redox potential	mV	<0.001	M–W	0.006	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Turbidity	NTU	<0.001	M–W	0.093	<0.001	M–W	<0.001	0.001	M–W	<0.001	<0.001	M–W	<0.001
Total suspended solids	mg/l	<0.001	M–W	0.962	<0.001	M–W	<0.001	0.008	M–W	<0.001	<0.001	M–W	<0.001
Electrical conductivity	µS/cm	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	0.070	<0.001	M–W	<0.001
Dissolved oxygen	mg/l	0.009	M–W	0.284	<0.001	M–W	0.001	<0.001	M–W	0.005	0.033	M–W	<0.001
Colour	Pa/Co	<0.001	M–W	0.006	<0.001	M–W	<0.001	0.001	M–W	0.007	0.001	M–W	<0.001
Temperature	°C	<0.001	M–W	0.863	<0.001	M–W	0.691	<0.001	M–W	0.063	<0.001	M–W	0.022
Biochemical oxygen demand	mg/l	0.001	M–W	0.054	<0.001	M–W	0.014	0.001	M–W	<0.001	<0.001	M–W	0.160
Chemical oxygen demand	mg/l	0.061	T–test	0.157	0.021	M–W	0.011	<0.001	M–W	0.926	0.007	M–W	0.030
Ammonia–nitrogen	mg/l	0.194	T–test	0.511	0.086	T–test	<0.001	<0.001	M–W	0.045	<0.001	M–W	0.873
Nitrate–nitrogen	mg/l	<0.001	M–W	0.019	<0.001	M–W	<0.001	0.004	M–W	<0.001	0.002	M–W	0.001
Ortho–phosphate–phosphorus	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Aluminium	mg/l	0.001	M–W	0.956	<0.001	M–W	<0.001	<0.001	M–W	0.386	<0.001	M–W	0.005
Boron	mg/l	0.020	M–W	0.019	0.102	T–test	0.188	<0.001	M–W	0.001	<0.001	M–W	<0.001
Calcium	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Cadmium	mg/l	0.002	M–W	0.004	<0.001	M–W	0.113	<0.001	M–W	0.516	<0.001	M–W	0.012
Chromium	mg/l	0.005	M–W	0.732	<0.001	M–W	0.212	<0.001	M–W	0.783	<0.001	M–W	0.738
Copper	mg/l	<0.001	M–W	0.971	<0.001	M–W	0.002	<0.001	M–W	0.020	<0.001	M–W	0.138
Iron	mg/l	0.001	M–W	0.432	<0.001	M–W	<0.001	0.004	M–W	0.016	<0.001	M–W	0.004
Potassium	mg/l	0.001	M–W	0.329	0.855	T–test	0.047	<0.001	M–W	0.505	<0.001	M–W	1.000
Magnesium	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Manganese	mg/l	<0.001	M–W	<0.001	<0.001	M–W	0.400	<0.001	M–W	0.152	<0.001	M–W	0.328
Sodium	mg/l	0.027	M–W	1.000	0.354	T–test	0.112	0.001	M–W	0.721	0.003	M–W	1.000
Nickel	mg/l	<0.001	M–W	0.818	<0.001	M–W	0.001	<0.001	M–W	0.001	<0.001	M–W	0.981
Zinc	mg/l	0.016	M–W	0.559	<0.001	M–W	0.042	0.020	M–W	0.205	0.002	M–W	0.409

Note: HC, high concentration; LC, low concentration; SGW, synthetic greywater; p value, significantly different, if $p < 0.05$ and not significantly different if $p > 0.05$; M–W, Mann–Whitney U–test; and NTU, nephelometric turbidity unit.

Table 6.7. (Continued).

(b) Effects of increasing contact time on remediation in ponds (P)

		HC–SGW						**LC–SGW**					
		Without ochre pellets			***With ochre pellets***			***Without ochre pellets***			***With ochre pellets***		
		P1 & P5			P2 & P6			P3 & P7			P4 & P8		
Parameter	**Unit**	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
pH	–	<0.001	M–W	0.751	<0.001	M–W	0.003	0.003	M–W	0.805	0.005	M–W	0.897
Redox potential	mV	<0.001	M–W	0.732	0.001	M–W	0.016	0.002	M–W	0.797	<0.001	M–W	0.747
Turbidity	NTU	0.318	T–test	0.405	<0.001	M–W	0.001	<0.001	M–W	0.030	0.001	M–W	0.533
Total suspended solids	mg/l	0.340	T–test	0.037	0.286	T–test	0.002	<0.001	M–W	0.009	0.001	M–W	0.235
Electrical conductivity	µS/cm	0.002	M–W	0.492	<0.001	M–W	0.139	0.275	T–test	0.197	<0.001	M–W	0.865
Dissolved oxygen	mg/l	0.008	M–W	0.012	<0.001	M–W	0.755	<0.001	M–W	<0.001	0.002	M–W	0.495
Colour	Pa/Co	0.513	T–test	0.071	0.049	M–W	0.463	0.177	T–test	0.063	0.312	T–test	0.093
Temperature	°C	<0.001	M–W	0.099	<0.001	M–W	0.668	<0.001	M–W	0.112	<0.001	M–W	0.549
Biochemical oxygen demand	mg/l	0.016	M–W	0.122	<0.001	M–W	0.037	<0.001	M–W	0.264	<0.001	M–W	0.092
Chemical oxygen demand	mg/l	<0.001	M–W	0.214	0.009	M–W	0.681	0.024	M–W	<0.001	0.069	T–test	0.009
Ammonia–nitrogen	mg/l	0.029	M–W	0.033	0.469	T–test	<0.001	0.107	T–test	0.303	<0.001	M–W	0.088
Nitrate–nitrogen	mg/l	0.001	M–W	0.005	<0.001	M–W	0.411	<0.001	M–W	0.028	<0.001	M–W	0.022
Ortho–phosphate–phosphorus	mg/l	0.001	M–W	0.025	0.003	M–W	0.194	<0.001	M–W	0.001	0.007	M–W	0.495
Aluminium	mg/l	0.329	T–test	0.020	0.001	M–W	0.921	<0.001	M–W	0.906	<0.001	M–W	0.951
Boron	mg/l	0.003	M–W	0.815	0.001	M–W	0.001	<0.001	M–W	0.001	<0.001	M–W	0.001
Calcium	mg/l	<0.001	M–W	<0.001	<0.001	M–W	0.406	0.001	M–W	0.002	<0.001	M–W	0.962
Cadmium	mg/l	0.002	M–W	0.006	<0.001	M–W	0.324	<0.001	M–W	0.243	<0.001	M–W	0.947
Chromium	mg/l	0.002	M–W	<0.001	<0.001	M–W	0.783	<0.001	M–W	0.557	<0.001	M–W	0.633
Copper	mg/l	<0.001	M–W	<0.001	<0.001	M–W	0.086	<0.001	M–W	0.630	<0.001	M–W	0.193
Iron	mg/l	<0.001	M–W	0.089	<0.001	M–W	0.009	<0.001	M–W	0.285	<0.001	M–W	0.054
Potassium	mg/l	0.016	M–W	<0.001	0.002	M–W	0.297	0.891	T–test	0.161	<0.001	M–W	1.000
Magnesium	mg/l	<0.001	M–W	<0.001	<0.001	M–W	0.894	<0.001	M–W	0.314	<0.001	M–W	0.049
Manganese	mg/l	<0.001	M–W	0.001	<0.001	M–W	0.018	<0.001	M–W	0.024	<0.001	M–W	0.979
Sodium	mg/l	0.009	M–W	0.267	0.019	M–W	0.664	0.044	M–W	0.441	0.004	M–W	0.920
Nickel	mg/l	<0.001	M–W	0.001	<0.001	M–W	0.560	<0.001	M–W	0.287	<0.001	M–W	0.002
Zinc	mg/l	<0.001	M–W	0.130	0.003	M–W	0.481	<0.001	M–W	0.057	<0.001	M–W	0.023

Note: HC, high concentration; LC, low concentration; SGW, synthetic greywater; p value, significantly different, if $p < 0.05$ and not significantly different if $p > 0.05$; M–W, Mann–Whitney U–test; and NTU, nephelometric turbidity unit.

Table 6.7. (Continued).

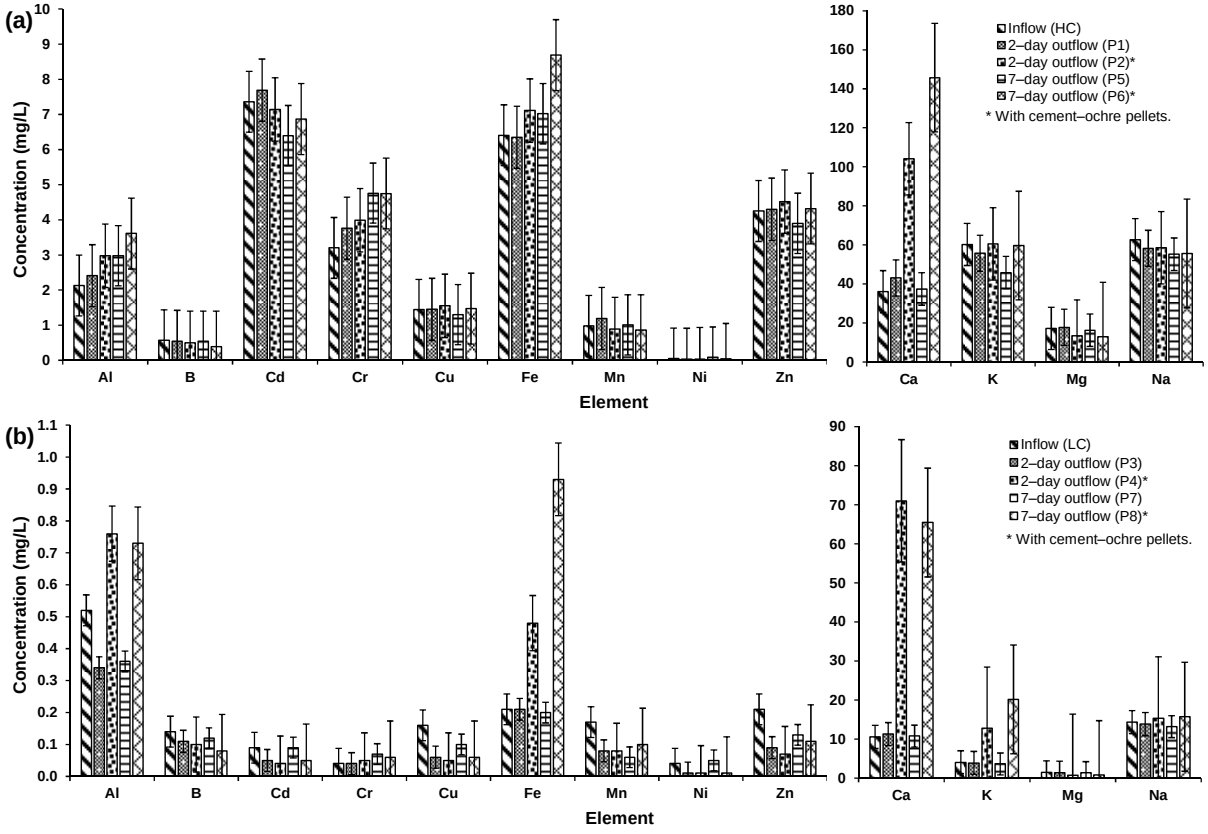
(c) Effects of pollutant loads on removals in ponds (P)

		2–day contact time						7–day contact time					
		Without ochre pellets			*With ochre pellets*			*Without ochre pellets*			*With ochre pellets*		
		P1 & P3			P2 & P4			P5 & P7			P6 & P8		
Parameter	**Unit**	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
Turbidity	NTU	<0.001	M–W	0.006	<0.001	M–W	0.066	<0.001	M–W	<0.001	<0.001	M–W	0.179
Total suspended solids	mg/l	<0.001	M–W	<0.001	<0.001	M–W	0.004	<0.001	M–W	<0.001	<0.001	M–W	0.248
Dissolved oxygen	mg/l	<0.001	M–W	0.069	0.002	M–W	0.405	0.655	T–test	0.047	0.435	T–test	0.372
Colour	Pa/Co	<0.001	M–W	<0.001	<0.001	M–W	0.327	<0.001	M–W	<0.001	<0.001	M–W	0.936
Biochemical oxygen demand	mg/l	<0.001	M–W	0.010	<0.001	M–W	0.006	<0.001	M–W	0.031	<0.001	M–W	0.726
Chemical oxygen demand	mg/l	0.018	M–W	0.032	0.001	M–W	0.232	0.001	M–W	<0.001	0.063	T–test	0.004
Ammonia–nitrogen	mg/l	<0.001	M–W	0.001	<0.001	M–W	0.406	<0.001	M–W	<0.001	<0.001	M–W	0.778
Nitrate–nitrogen	mg/l	<0.001	M–W	0.826	<0.001	M–W	0.006	<0.001	M–W	0.524	<0.001	M–W	<0.001
Ortho–phosphate–phosphorus	mg/l	<0.001	M–W	0.664	0.001	M–W	0.001	<0.001	M–W	0.001	<0.001	M–W	<0.001
Aluminium	mg/l	<0.001	M–W	0.213	<0.001	M–W	0.014	<0.001	M–W	0.064	<0.001	M–W	0.458
Boron	mg/l	<0.001	M–W	0.035	0.020	M–W	0.290	<0.001	M–W	0.038	0.024	M–W	0.152
Calcium	mg/l	<0.001	M–W	<0.001	0.001	M–W	<0.001	<0.001	M–W	0.265	<0.001	M–W	0.005
Cadmium	mg/l	0.003	M–W	<0.001	0.003	M–W	<0.001	<0.001	M–W	0.201	<0.001	M–W	0.076
Chromium	mg/l	0.004	M–W	0.039	<0.001	M–W	0.054	<0.001	M–W	0.274	<0.001	M–W	0.759
Copper	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Iron	mg/l	<0.001	M–W	0.221	0.025	M–W	0.019	<0.001	M–W	0.003	<0.001	M–W	0.043
Potassium	mg/l	0.002	M–W	0.729	0.029	M–W	0.488	0.103	T–test	0.001	<0.001	M–W	1.000
Magnesium	mg/l	0.016	M–W	0.005	<0.001	M–W	<0.001	0.171	T–test	0.866	<0.001	M–W	0.003
Manganese	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	0.955
Sodium	mg/l	0.026	M–W	0.488	<0.001	M–W	0.106	0.009	M–W	0.009	<0.001	M–W	0.462
Nickel	mg/l	<0.001	M–W	<0.001	<0.001	M–W	0.149	<0.001	M–W	<0.001	<0.001	M–W	0.006
Zinc	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	0.398	<0.001	M–W	0.426

Note: HC, high concentration; LC, low concentration; SGW, synthetic greywater; p value, significantly different, if $p < 0.05$ and not significantly different if $p > 0.05$; M–W, Mann–Whitney U–test; NTU, nephelometric turbidity unit.

6.4.2. Minerals and Trace Elements in Greywater

The chemical analysis of trace element concentrations of treated outflow SGW are summarised in Table 6.5. A statistical analysis of significant differences between inflow and outflow water qualities showed that concentrations of B, Mg and Mn were removed significantly ($p < 0.05$) from treated HC–SGW with ochre pellets. However, concentrations of Ca, Fe and Cu (for 2–day contact time) at the presence of ochre pellets were significantly higher in the outflow than those concentrations in the HC–SGW inflow. No significant ($p > 0.05$) changes in concentrations for the elements Al, Cd, Cr, Ni and Zn were noted (Table 6.6 and Figure 6.4–a).



Note: Al, aluminium; B, boron; Cd, cadmium; Cr, chromium; Cu, copper; Fe, iron; Mn; manganese; Ni, nickel; Zn, zinc; Ca, calcium; K, potassium; Mg, magnesium; and Na, sodium.

Figure 6.4. Comparison between inflow and outflow synthetic greywater treated by various experimental set–up designs of simulated stabilisation ponds (P) in terms of trace element concentrations of; (a) high concentration synthetic greywater (HC–SGW); and (b) low concentration synthetic greywater (LC–SGW).

Concerning cement–ochre pellets at the end of experiment, significant ($p < 0.05$) accumulations of Cd, Cr, B, Ni, Mg, Cu, Zn and Mn gave an indication of remediation concerning SGW for different treatment systems using ochre pellets at an early stage of treatment. Therefore, after successive times of treatment, the ochre pellets reached their adsorption capacity in terms of these elements, except for B, Mg and Mn with respect to the treatment of HC–SGW. For the treatment of LC–SGW with ochre pellets, the concentrations of B, Cd, Cu, Mg, Mn, Ni and Zn were significantly lower than those measured for the inflow. However, Al, Ca, Cr and Fe increased significantly ($p < 0.05$) in the outflow compared with those concentrations in the inflow. No significant ($p > 0.05$) differences in K and Na concentrations were noted (Table 6.6 and Figure 6.4–b).

In both types of SGW treated with ochre pellets, Ca concentrations in outflows were the highest among all elements and higher than those in the other outflows. Due to the addition of cement in pelletised ochre, Ca release from ochre pellets into greywater increased significantly (Table 6.2 and Figure 6.2). Since there was a positive correlation ($r = 0.499$, $p < 0.001$) between Ca and pH in the outflows, it is expected that there is no mobilisation of Fe and Al from ochre pellets to the treated greywater. This should particularly be the case for HC–SGW under high pH conditions (Carr, 2012).

The presence of cement–ochre pellets in ponds P4 and P8 treating LC–SGW significantly ($p < 0.05$) impacted on the concentrations of Mg, Cu (after two days of contact time only), B (7–days) and Cd (7–days) in the outflow compared with these elements in the outflow of ponds P3 and P7, where ochre pellets were absent. However, significant increases of concentrations in LC–SGW outflow in terms of Al, Ca, Fe, K (2–days), Ni (2–days) and Zn (2–days) were noted. However, no significant ($p < 0.05$) differences in Cr, Mn and Na were detected (Tables 6.6 and 6.7). For the treatment of HC–SGW, the concentrations of B, Mg, Cd (2–days), Mn (2–days) and Ni (7–days) were

significantly decreased by the presence of ochre in the treatment by ponds P2 and P6 compared with the outflow of ponds P1 and P5, where ochre pellets were absent. Furthermore, Al, Cr, K, Na and Zn concentrations did not change significantly ($p > 0.05$).

In addition, an increase in contact time for the treatment ponds with ochre pellets significantly ($p < 0.05$) reduced B and Mn (for HC–SGW), while no significant ($p > 0.05$) effects on the concentrations of Al, Ca, Cd, Cr, Cu, K, Na, Fe (for LC–SGW), Mg (for HC–SGW), Ni (for HC–SGW) and Zn (for HC–SGW) were computed. However, trace element concentrations in the outflow depend on the behaviour of ochre pellets for adsorption. Furthermore, system performance can vary due to the interaction between the operational variables of the experiment such as the contact time between the ochre pellets and the greywater, and the pollutant strength of loading, regardless of the effect of the outdoor environment. The mobilisation of Fe and Al minerals from ochre pellets due to the decrease in redox potential allows for phosphorus to take some vacant places on and within the ochre pellets (Dobbie et al., 2009).

No significant ($p > 0.05$) differences in the removals of B, Cr, K and Na for both HC–SGW and LC–SGW at the presence of ochre pellets were computed. However, the removals of Ca, Fe and Al (2–days) were significantly ($p < 0.05$) high for the treatment of HC–SGW compared with the removals linked to the treatment of LC–SGW. Elements such as Cu, Mg Cd (2–days), Mn (2–days), Zn (2–days) and Ni (7–days) were significantly ($p < 0.05$) removed for the treatment of LC–SGW using ochre pellets compared to HC–SGW (Table 6.7). Since the same quantity of cement–ochre pellets was applied in each treatment, the removal efficiency depends on the initial concentrations of trace elements in greywater. Therefore, higher removals can be expected for greywater treatment systems, where low concentrations of trace elements are presented (Sahoo et al., 2014).

6.5 Accumulation of Trace Elements in Ochre Pellets

At the end of treatments involving HC–SGW and LC–SGW, the accumulation of trace elements (Table 6.9) in cement–ochre pellets was assessed (Table 6.2). Results show that all elements (except for Ca) were significantly ($p < 0.05$) adsorbed by ochre pellets after treating both types of the synthetic greywater. This was particularly the case for HC–SGW (Table 6.9). The ochre adsorption capacity of trace elements at 2–day contact time for the treatment of HC–SGW in pond P2 follows the rank relationship Cd > Cr > B > Ni > Mg > Cu > Zn > Mn > Al > Fe > Ca. In comparison, the relationship for pond P6 at 7–day contact time was Cd > B > Cr > Ni > Cu > Mg > Zn > Al > Mn > Fe > Ca (Figure 6.5).

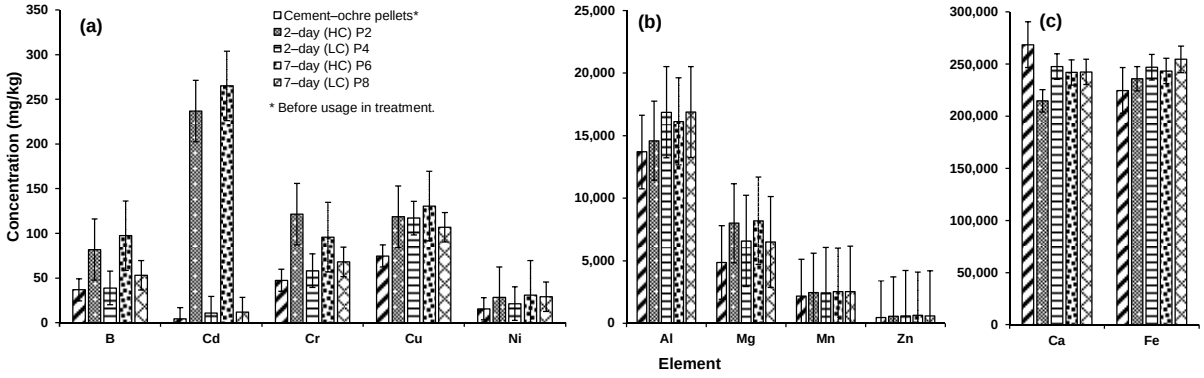


Figure 6.5. Comparison between the adsorbed trace element concentrations by cement–ochre pellets treating synthetic greywater in various experimental set–up designs of simulated stabilisation ponds (P): *(a)* boron (B), cadmium (Cd), chromium (Cr), copper (Cu) and nickel (Ni); *(b)* aluminium (Al), magnesium (Mg), manganese (Mn) and zinc (Zn); and *(c)* calcium (Ca) and iron (Fe).

Table 6.8. Trace elements (mg/kg) accumulated in cement–ochre pellets after the treatment period in mesocosm–scale stabilisation ponds (P) treating (a) high concentration synthetic greywater (HC–SGW); and (b) low concentration synthetic greywater (LC–SGW).

(a)							**HC–SGW**					
	Cement–ochre pellets in P2						***Cement–ochre pellets in P6***					
Element	n	Mean	SD	Min	Max	Accumulation (%)	n	Mean	SD	Min	Max	Accumulation (%)
Aluminium	18	14589.7	1153.27	12352.2	16197.7	6.5	18	16130.6	969.31	14621.8	17984.0	17.8
Boron	15	82.0	17.80	55.5	115.9	122.1	15	97.4	15.84	79.2	124.4	163.7
Calcium	21	214795.4	12783.36	190833.2	231786.0	-20.0	21	242023.8	11280.09	217711.6	258784.8	-9.9
Cadmium	14	237.0	89.84	26.4	334.2	5208.6	12	265.1	50.32	169.8	356.4	5838.1
Chromium	27	121.5	74.45	41.3	277.4	155.6	27	95.8	34.21	51.5	197.2	101.5
Copper	27	118.7	69.27	52.8	279.8	58.7	27	130.5	80.23	42.4	329.5	74.5
Iron	24	235900.8	18729.34	198789.6	263774.4	5.0	24	243388.2	16023.00	210822.0	265205.2	8.4
Magnesium	27	7999.2	755.80	5994.7	8759.9	64.6	27	8194.8	712.29	6240.2	8976.9	68.7
Manganese	27	2443.6	230.23	2024.0	2894.8	12.5	27	2522.1	255.90	2104.0	3050.9	16.1
Nickel	21	28.2	18.74	2.4	58.1	81.8	20	30.7	20.60	2.3	73.2	97.8
Zinc	24	546.0	160.59	334.6	917.9	22.0	24	626.7	189.38	441.2	1032.5	40.0
(b)							**LC–SGW**					
	Cement–ochre pellets in P4						***Cement–ochre pellets in P8***					
Element	n	Mean	SD	Min	Max	Accumulation (%)	n	Mean	SD	Min	Max	Accumulation (%)
Aluminium (Al)	18	16867.7	1114.87	14944.1	18942.3	23.2	18	16874.5	914.04	15309.2	18198.2	23.2
Boron (B)	15	38.9	16.62	22.5	69.5	5.3	15	53.1	17.16	35.4	82.3	43.9
Calcium (Ca)	21	247556.5	15332.50	220230.0	276971.6	-7.8	21	242354.2	9066.79	224942.0	256550.4	-9.7
Cadmium (Cd)	15	10.7	21.03	0.0	82.7	139.6	18	11.8	10.14	0.0	26.2	164.3
Chromium (Cr)	27	58.4	25.40	23.8	110.2	22.8	27	68.1	33.26	29.5	153.0	43.2
Copper (Cu)	27	117.1	84.00	17.6	319.4	56.6	27	106.9	78.81	40.0	288.2	42.9
Iron (Fe)	24	246941.2	17821.97	210530.0	273850.4	9.9	24	254510.0	13227.47	230163.2	274306.4	13.3
Magnesium (Mg)	27	6586.0	572.12	5121.9	7399.4	35.5	27	6492.4	511.48	5170.8	6962.6	33.6
Manganese (Mn)	27	2424.3	168.58	2160.8	2839.4	11.6	27	2529.9	144.24	2302.0	2855.0	16.5
Nickel (Ni)	14	21.3	12.16	0.1	43.9	37.1	17	29.0	20.60	0.4	59.2	86.6
Zinc (Zn)	24	589.0	188.00	436.6	1059.1	31.5	24	578.2	175.57	449.2	1013.4	29.1

Note: P2 and P4 treat greywaters with a 2–day contact time; P6 and P8 treat greywaters with a 7–day contact time; n, number of tested samples; SD, standard deviation; Min, minimum; and Max, maximum.

However, no significant ($p > 0.05$) differences for the concentrations of Ni, B (2–days), Cd (2–days) and Cr (2–days) were found, if these elements are compared with those corresponding concentrations in ochre pellets before treating LC–SGW (Table 6.9 and Figure 6.5). In general, accumulated elements in ochre pellets treating LC–SGW were ranked for pond P4 at 2–day contact time as follows: Cd > Cu > Ni > Mg > Zn > Al > Cr > Mn > Fe > B > Ca. In comparison, at 7–day contact time for pond P8, the relationship was as shown here: Cd > Ni > B > Cr > Cu > Mg > Zn > Al > Mn > Fe > Ca. The Ca content decreased significantly in ochre pellets treating both types of greywaters due to its mobilisation into greywater (Figure 6.6). Correspondingly, Ca concentrations released into greywater raised significantly ($p < 0.05$), as indicated in Tables 6.5, 6.6 and as well as Figure 6.4.

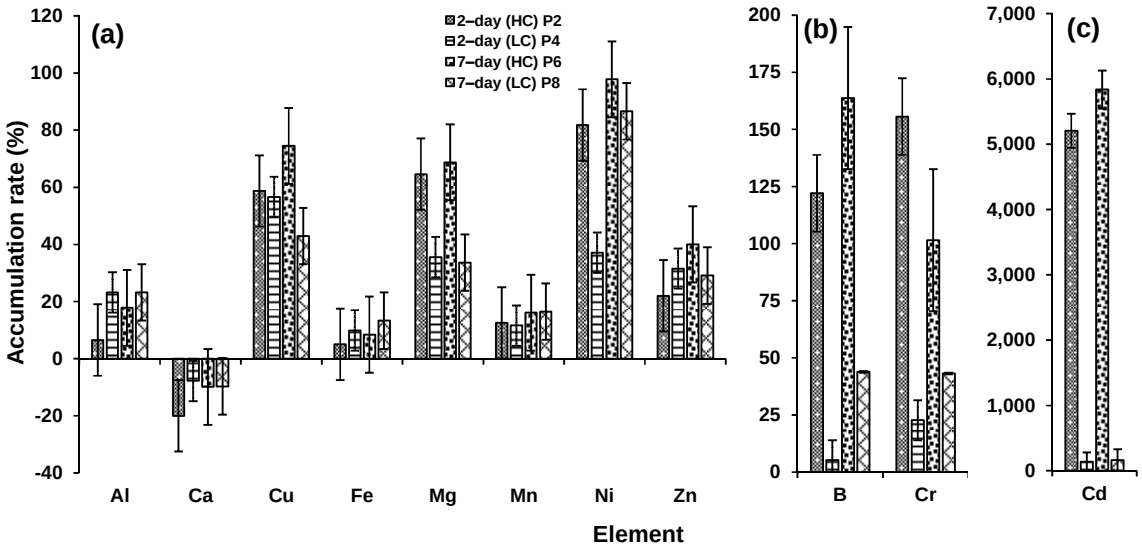


Figure 6.6. Comparison between the accumulative rates of trace element concentrations in cement–ochre pellets treating synthetic greywater in various experimental set–up designs of simulated stabilisation ponds (P): *(a)* aluminium (Al), calcium (Ca), copper (Cu), iron (Fe), magnesium (Mg), manganese (Mn), nickel (Ni) and zinc (Zn); *(b)* boron (B) and chromium (Cr); and *(c)* cadmium (Cd).

Some element concentrations increased in the outflows of the treatment systems without the presence of ochre pellets, since all systems were subjected to the impacts of natural forces such as precipitation. It is also likely that contaminants such as dust particles and plant leaves entered the ponds. However, all assessed elements originating either from greywater or from the environment such as B, Cd, Mg, Mn, Ni and Zn were significantly ($p < 0.05$) adsorbed by cement–ochre pellets without mobilisation. Furthermore, the outflows of systems with ochre pellets also showed significant ($p < 0.05$) increases in concentrations of Al, Cr, Fe, Cu (treatment of HC–SGW), K (treatment of LC–SGW) and Na (treatment of LC–SGW). On the other hand, Al, Cr, Fe, Cu, K and Na concentrations in ochre pellets did not decrease (Table 6.8 and Figure 6.6). Therefore, it is likely that elements were remobilised back into the water column after adsorption.

In addition, an increase in contact time significantly ($p < 0.05$) impacted on the rise in concentrations of the elements Al, B, Ca and Zn for the treatment of HC–SGW, and B and Mn for the treatment of LC–SGW, which accumulated in cement–ochre pellets compared with those concentrations in ochre pellets for a 2–day contact time of treatment. However, no significant ($p > 0.05$) effect of increased contact time was observed when studying the accumulation of other elements (Table 6.9). Furthermore, insignificant ($p > 0.05$) decreases in the content of some elements such as Ca, Cu, Mg and Zn in ochre pellets for the treatment of LC–SGW and Cr for the corresponding treatment of HC–SGW were computed for increasing contact time of treatment compared with those concentrations in ochre pellets for systems operating at 2–day contact time (Table 6.8 and Figure 6.5).

The effect of pollutant loads in SGW on trace element accumulations in ochre pellets showed that the concentrations of accumulated Al, Fe, Ca (2–days) and Zn (2–days) were significantly ($p < 0.05$) high in ochre pellets treating LC–SGW. In

comparison, the concentrations of B, Cd, Cr, Mg and Zn (7–days) were significantly ($p < 0.05$) higher in ochre pellets treating HC–SGW compared to LC–SGW (Table 6.5 and Figure 6.6). It follows that the amount of adsorbed elements correlated well with the corresponding initial trace element concentrations in greywater. Therefore, a high remediation of an element at low concentration of greywater indicates sufficient availability of vacant places on or within ochre pellets to sustain the adsorbed metals (Sahoo et al., 2014).

Table 6.9. Statistical analysis to assess (a) differences between the trace elements (mg/kg) accumulated in cement–ochre pellets before and after treatment; and (b) the effects of increasing contact time and pollutant loads on trace element (mg/kg) accumulations in cement–ochre pellets.

(a) Comparison of accumulative elements in cement–ochre pellets before and after the treatment period within the ponds (P)

	2–day contact time						7–day contact time					
	HC–SGW (P2)			*LC–SGW (P4)*			*HC–SGW (P6)*			*LC–SGW (P8)*		
Element	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
Aluminium	0.546	T–test	0.009	0.089	T–test	<0.001	0.386	T–test	<0.001	0.022	M–W	<0.001
Boron	0.290	T–test	<0.001	0.014	M–W	0.520	0.053	T–test	<0.001	0.116	T–test	0.013
Calcium	0.016	M–W	<0.001	0.771	T–test	<0.001	0.908	T–test	<0.001	0.604	T–test	<0.001
Cadmium	<0.001	M–W	<0.001	<0.001	M–W	0.526	<0.001	M–W	<0.001	<0.001	M–W	0.039
Chromium	<0.001	M–W	<0.001	<0.001	M–W	0.082	0.002	M–W	<0.001	<0.001	M–W	0.009
Copper	<0.001	M–W	0.003	<0.001	M–W	0.019	<0.001	M–W	0.002	<0.001	M–W	0.014
Iron	0.374	T–test	0.013	0.041	M–W	<0.001	0.062	T–test	<0.001	0.087	T–test	<0.001
Magnesium	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Manganese	0.011	M–W	<0.001	0.190	T–test	<0.001	0.003	M–W	<0.001	0.140	T–test	<0.001
Nickel	0.008	M–W	0.044	0.462	T–test	0.166	0.026	M–W	0.019	0.019	M–W	0.071
Zinc	<0.001	M–W	0.002	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001

(b) Effects of increasing contact time and pollutant loads on ponds (P)

	Effects of increasing contact time						Effects of pollutant loads					
	HC–SGW (P2 and P6)			*LC–SGW (P4 and P8)*			*2–day contact time (P2 and P4)*			*7–day contact time (P6 and P8)*		
Element	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
Aluminium	0.917	T–test	<0.001	0.750	T–test	0.984	0.980	T–test	<0.001	0.287	T–test	0.024
Boron	0.603	T–test	0.019	0.033	M–W	0.017	0.116	T–test	<0.001	0.069	T–test	<0.001
Calcium	0.438	T–test	<0.001	0.560	T–test	0.190	0.807	T–test	<0.001	0.479	T–test	0.917
Cadmium	0.003	M–W	0.643	<0.001	M–W	0.179	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Chromium	<0.001	M–W	0.856	0.002	M–W	0.223	<0.001	M–W	0.002	<0.001	M–W	0.005
Copper	<0.001	M–W	0.586	<0.001	M–W	0.762	<0.001	M–W	0.452	<0.001	M–W	0.054
Iron	0.064	T–test	0.144	0.020	M–W	0.167	0.214	T–test	0.042	0.031	M–W	0.016
Magnesium	<0.001	M–W	0.210	<0.001	M–W	0.598	0.030	M–W	<0.001	0.002	M–W	<0.001
Manganese	0.366	T–test	0.242	0.356	T–test	0.017	0.941	T–test	0.726	0.573	T–test	0.891
Nickel	0.018	M–W	0.754	0.196	T–test	0.208	0.104	T–test	0.510	0.052	T–test	0.800
Zinc	<0.001	M–W	0.002	<0.001	M–W	0.635	<0.001	M–W	0.099	<0.001	M–W	0.021

Note: M–W, Mann–Whitney U–test; p value, significantly difference, if $p < 0.05$ and not significantly different, if $p > 0.05$.

Chapter Seven

Cement–Ochre Pellets with Floating Treatment Wetland Systems for Synthetic Greywater Remediation

Abed S.N., Almuktar S.A.A.A.N, Scholz, M. Performance of Combined Cement–ochre Pellet and Floating Wetland Treatment Systems for Synthetic Greywater Remediation. Ecological Engineering, *(submitted)*.

7.1. Overview

In this chapter, the test results carried out from the floating treatment wetlands experiment are grouped into sets according to their experimental set–up design to achieve the main aim and objective (4), section 1.5. The results are discussed in each of the following sections to evaluate the effects of the operational variable parameters such as concentrations of contaminations, contact time of treatment, presence of vegetation, and presence of cement–ochre solids on the performance of each system. Focusing on a combination of floating *P. australis* with cement–ochre pellets, the improvement of greywater quality in treatment systems, in particular removal of organic matter, nutrients, and trace elements, was assessed. Furthermore, the concentrations of trace elements accumulated in ochre pellets and the *P. australis* tissues were analysed at end of the experiment and discussed.

7.2. Inflow and outflow water quality

Table 7.1 illustrates the inflow water quality parameters of both the low (LC) and high (HC) pollutant concentrations of the synthetic greywater (SGW). Moreover, outflow greywater quality values, which were collected after treatment of SGW in different experimental design set–ups, are shown in Table 7.2. Outflow quality parameters of control wetlands, which received tap water, are also included in Table 7.2.

Table 7.1. Physiochemical characteristics of inflow synthetic greywater (SGW).

Parameter	***Unit***	***Inflow (low concentration)***					***Inflow (high concentration)***				
		n	Mean	SD	Min	Max	n	Mean	SD	Min	Max
pH	–	81	6.9	0.48	5.3	7.9	81	8.4	1.61	5.4	11.5
Redox potential	mV	81	34.1	21.23	-18.1	111.2	81	-36.6	74.22	-182.1	97.9
Turbidity	NTU	81	22.9	7.14	9.8	41.6	81	188.9	47.22	18.3	308.0
Total suspended solids	mg/l	81	39.9	15.94	10.0	87.0	81	317.0	58.35	173.0	473.0
Electrical conductivity	µS/cm	81	164.6	63.24	98.7	452.0	81	988.5	196.09	612.0	1677.0
Dissolved oxygen	mg/l	81	10.4	1.24	7.7	12.3	81	10.5	1.39	6.9	12.6
Colour	Pa/Co	81	214.5	64.07	26.0	340.0	81	1587.8	379.89	787.0	2499.0
Temperature	°C	81	17.7	4.58	6.7	27.0	81	16.9	5.40	6.5	27.8
Biochemical oxygen demand	mg/l	81	17.6	8.00	2.0	40.0	81	34.7	12.99	10.0	60.0
Chemical oxygen demand	mg/l	81	28.9	14.47	8.2	86.7	81	129.2	34.68	63.9	221.0
Ammonia–nitrogen	mg/l	81	0.2	0.22	0.0	1.1	81	0.4	0.19	0.1	1.1
Nitrate–nitrogen	mg/l	81	1.3	1.21	0.1	7.6	81	8.9	6.38	0.2	29.8
Ortho–phosphate–phosphorus	mg/l	81	8.4	4.36	3.3	27.4	81	59.1	14.16	30.6	94.2
Element											
Aluminium (Al)	mg/l	45	0.52	0.528	0.09	1.56	45	2.13	0.869	0.76	4.77
Boron (B)	mg/l	33	0.14	0.067	0.10	0.36	33	0.57	0.068	0.49	0.72
Calcium (Ca)	mg/l	55	10.54	0.853	8.51	11.81	45	36.08	8.750	22.38	50.72
Cadmium (Cd)	mg/l	42	0.09	0.056	0.04	0.23	39	7.36	2.981	3.67	11.58
Chromium (Cr)	mg/l	54	0.04	0.063	0.01	0.23	54	3.20	0.918	1.67	5.70
Copper (Cu)	mg/l	63	0.16	0.058	0.10	0.30	63	1.44	0.435	0.95	2.03
Iron (Fe)	mg/l	48	0.21	0.102	0.11	0.43	48	6.41	2.476	2.48	10.37
Potassium (K)	mg/l	12	4.04	0.448	3.41	4.65	12	60.16	1.684	58.31	62.53
Magnesium (Mg)	mg/l	48	1.45	0.191	1.19	1.78	48	17.16	2.119	12.68	20.33
Manganese (Mn)	mg/l	63	0.17	0.084	0.07	0.26	63	0.98	0.257	0.64	1.29
Sodium (Na)	mg/l	12	14.32	1.662	12.01	16.00	12	62.68	14.538	45.50	76.92
Nickel (Ni)	mg/l	51	0.04	0.065	0.01	0.18	51	0.05	0.065	0.01	0.20
Zinc (Zn)	mg/l	39	0.21	0.159	0.01	0.39	39	4.25	1.500	2.22	6.34

Note: n, number of tested samples; SD, standard deviation; Min, minimum; Max, maximum; and NTU, nephelometric turbidity unit.

Table 7.2. Outflow quality of the synthetic greywater (SGW) treated for 2– and 7–day contact time; a) high concentration greywater (HC–SGW) at 2–day contact time, b) low concentration greywater (LC–SGW) at 2–day contact time, c) high concentration greywater (HC–SGW) at 7–day contact time, and d) low concentration greywater (LC–SGW) at 7–day contact time.

a) HC–SGW (2–day contact time)

			2–day outflow (HC)–T1						*2–day outflow (HC)–T2*				
Parameter	***Unit***	n	Mean	SD	Min	Max	Rem (%)	n	Mean	SD	Min	Max	Rem (%)
pH	–	85	7.4	1.09	5.7	9.5	na	36	8.8	1.69	6.6	12.5	na
Redox potential	mV	85	8.1	52.68	-89.5	87.2	na	36	-54.8	83.66	-227.7	56.7	na
Turbidity	NTU	85	175.9	59.61	39.8	319.0	6.9	36	223.8	97.40	88.9	434.0	-18.5
Total suspended solids	mg/l	85	302.9	75.19	79.0	455.0	4.4	36	422.5	152.77	178.0	662.0	-33.3
Electrical conductivity	µS/cm	85	987.4	107.25	713.0	1225.0	na	36	1174.5	282.81	766.0	1924.0	na
Dissolved oxygen	mg/l	85	9.0	1.03	5.5	11.9	14.3	36	9.0	1.24	6.5	12.2	14.3
Colour	Pa/Co	85	1525.6	411.54	455.0	2361.0	3.9	36	2150.8	864.04	784.0	4175.0	-35.5
Temperature	°C	85	17.1	4.92	5.8	24.6	na	36	17.4	4.87	6.0	23.8	na
Biochemical oxygen demand	mg/l	85	17.7	6.40	0.0	30.0	49.0	36	11.1	5.89	0.0	28.0	68.0
Chemical oxygen demand	mg/l	85	96.3	32.01	33.8	172.0	25.5	36	109.2	24.38	56.9	151.0	15.5
Ammonia–nitrogen	mg/l	85	0.4	0.21	0.0	1.2	0.0	36	0.4	0.13	0.2	0.7	0.0
Nitrate–nitrogen	mg/l	85	14.1	6.40	0.2	32.0	-58.4	36	14.3	5.02	4.7	26.5	-60.7
Ortho–phosphate–phosphorus	mg/l	85	52.0	14.87	15.8	95.5	12.0	36	21.1	5.81	6.9	29.2	64.3
Element													
Aluminium	mg/l	39	1.54	1.479	0.30	5.04	27.70	39	2.02	1.624	0.48	4.94	5.16
Boron	mg/l	35	0.53	0.086	0.43	0.73	7.02	35	0.41	0.079	0.34	0.57	28.07
Calcium	mg/l	37	42.50	4.561	34.22	49.76	-17.79	37	81.39	23.641	46.90	115.66	-125.58
Cadmium	mg/l	42	4.90	2.730	1.55	8.93	33.42	42	4.10	1.839	1.79	7.25	44.36
Chromium	mg/l	58	2.48	2.060	0.59	6.69	22.50	58	2.74	2.021	0.83	5.85	14.38
Copper	mg/l	63	0.95	0.561	0.29	1.57	34.03	63	0.90	0.375	0.44	1.36	37.50
Iron	mg/l	51	4.31	2.928	1.08	9.02	32.76	51	4.71	2.744	1.37	8.10	26.52
Potassium	mg/l	14	52.79	1.322	50.41	54.90	12.25	14	54.03	11.214	44.74	71.52	10.19
Magnesium	mg/l	48	17.32	1.296	14.49	20.30	-0.93	48	11.01	2.533	7.57	14.16	35.84
Manganese	mg/l	63	0.48	0.320	0.10	0.91	51.02	63	0.51	0.255	0.29	0.97	47.96
Sodium	mg/l	14	58.54	11.080	42.44	73.63	6.60	14	56.95	9.494	42.01	67.61	6.18
Nickel	mg/l	53	0.02	0.019	0.00	0.06	60.00	53	0.02	0.019	0.00	0.08	60.00
Zinc	mg/l	44	2.86	1.680	0.70	4.70	32.71	44	2.58	1.114	1.01	4.18	39.29
			2–day outflow (HC)–T3						*2–day outflow (HC)–T4*				
pH	–	85	7.8	1.37	5.6	9.8	na	36	8.7	1.73	6.6	12.7	na
Redox potential	mV	85	-3.0	62.95	-107.6	88.6	na	36	-49.9	83.61	-232.8	42.1	na
Turbidity	NTU	85	192.1	50.87	102.0	341.0	-1.7	36	191.3	84.41	106.0	456.0	-1.3
Total suspended solids	mg/l	85	321.8	56.68	165.0	447.0	-1.5	36	337.4	109.45	161.0	661.0	-6.4
Electrical conductivity	µS/cm	85	965.2	106.68	627.0	1208.0	na	36	1178.4	264.41	806.0	1944.0	na
Dissolved oxygen	mg/l	85	10.2	0.73	8.7	12.1	2.9	36	10.0	0.52	8.9	10.9	4.8
Colour	Pa/Co	85	1527.6	326.28	677.0	2311.0	3.8	36	1935.6	702.18	702.0	3438.0	-21.9
Temperature	°C	85	17.1	4.75	6.1	23.6	na	36	17.2	4.73	6.0	23.4	na
Biochemical oxygen demand	mg/l	85	14.7	7.78	0.0	40.0	57.6	36	11.7	7.71	0.0	35.0	66.3
Chemical oxygen demand	mg/l	85	106.6	22.68	43.3	164.0	17.5	36	100.3	21.08	41.7	131.0	22.4
Ammonia–nitrogen	mg/l	85	0.4	0.16	0.0	0.9	0.0	36	0.4	0.09	0.2	0.6	0.0
Nitrate–nitrogen	mg/l	85	9.4	4.67	0.5	24.0	-5.6	36	12.9	7.03	2.4	26.9	-44.9
Ortho–phosphate–phosphorus	mg/l	85	46.2	10.74	23.7	70.1	21.8	36	19.5	4.98	9.0	30.3	67.0
Element													
Aluminium	mg/l	39	2.41	1.016	0.74	4.25	-13.15	39	2.98	2.087	0.84	7.14	-39.91
Boron	mg/l	35	0.54	0.060	0.42	0.66	5.26	35	0.50	0.078	0.40	0.68	12.28
Calcium	mg/l	37	43.02	2.411	35.94	46.28	-19.24	37	104.13	32.868	46.28	141.65	-188.61
Cadmium	mg/l	42	7.69	1.064	4.95	8.98	-4.48	42	7.14	2.429	4.23	11.75	2.99
Chromium	mg/l	58	3.76	1.203	1.34	4.98	-17.5	58	3.99	1.806	1.87	7.03	-24.69
Copper	mg/l	63	1.45	0.113	1.28	1.70	-0.69	63	1.55	0.308	1.23	2.09	-7.64
Iron	mg/l	51	6.35	2.423	1.56	9.29	0.94	51	7.11	2.934	2.60	11.55	-10.92
Potassium	mg/l	14	55.68	4.486	49.48	60.69	7.45	14	60.47	15.561	49.85	85.46	-0.52
Magnesium	mg/l	48	17.76	1.392	13.92	19.55	-0.20	48	13.33	4.526	7.53	19.80	22.32
Manganese	mg/l	63	1.19	0.063	1.06	1.29	-21.4	63	0.89	0.396	0.61	1.59	9.18
Sodium	mg/l	14	58.19	10.620	42.35	68.22	7.16	14	58.54	11.630	41.37	76.33	6.60
Nickel	mg/l	53	0.03	0.018	0.00	0.06	40.00	53	0.03	0.033	0.00	0.10	40.00
Zinc	mg/l	44	4.30	0.524	3.12	5.25	-1.42	44	4.52	0.961	2.90	6.40	-6.35

Note: HC, high pollutant concentrations; T1, treatment system with only *P. australis*; T2, treatment system with *P. australis* and ochre pellets; T3, treatment system without *P. australis* or ochre pellets; T4, treatment system with only ochre pellets; n, number of tested samples; SD, standard deviation; Min, minimum; Max, maximum; Rem, removal; NTU, nephelometric turbidity unit; na, not applicable.

Table 7.2. (Continued).

b) LC–SGW (2–day contact time)

		2–day outflow (LC)–T5						*2–day outflow (LC)–T6*					
Parameter	***Unit***	n	Mean	SD	Min	Max	Rem (%)	n	Mean	SD	Min	Max	Rem (%)
pH	–	85	7.0	0.71	6.1	10.2	na	36	10.5	1.12	8.0	12.5	na
Redox potential	mV	85	27.5	32.18	-120.8	72.4	na	36	-137.4	54.91	-224.8	-15.0	na
Turbidity	NTU	85	28.2	37.09	3.4	262.0	-23.1	36	39.2	45.10	9.8	260.0	-71.2
Total suspended solids	mg/l	85	41.7	43.57	8.0	316.0	-4.5	36	62.0	49.93	13.0	262.0	-55.4
Electrical conductivity	µS/cm	85	145.9	30.41	15.5	325.0	na	36	371.5	260.12	4.1	1307.0	na
Dissolved oxygen	mg/l	85	9.3	1.08	6.9	12.5	10.6	36	8.8	0.87	6.5	10.6	15.4
Colour	Pa/Co	85	183.7	74.89	73.0	476.0	14.4	36	308.2	134.65	103.0	683.0	-43.7
Temperature	°C	85	17.0	4.84	6.0	23.0	na	36	16.6	4.55	6.2	22.9	na
Biochemical oxygen demand	mg/l	85	9.9	5.49	0.0	30.0	43.8	36	5.4	4.36	0.0	18.0	69.3
Chemical oxygen demand	mg/l	85	32.4	14.55	10.6	89.6	-13.8	36	29.6	16.67	9.6	73.4	-2.4
Ammonia–nitrogen	mg/l	85	0.1	0.07	0.0	0.3	50.0	36	0.2	0.14	0.0	0.5	0.0
Nitrate–nitrogen	mg/l	85	1.7	1.13	0.0	5.8	-30.8	36	0.4	0.33	0.0	1.6	69.2
Ortho–phosphate–phosphorus	mg/l	85	7.6	3.90	3.2	120	9.5	36	3.2	1.16	1.5	6.4	86.2
Element													
Aluminium	mg/l	39	0.08	0.054	0.01	0.19	84.62	39	1.07	0.874	0.08	2.54	-105.77
Boron	mg/l	35	0.11	0.010	0.10	0.14	21.43	35	0.09	0.011	0.08	0.11	35.71
Calcium	mg/l	46	11.51	0.926	10.01	13.18	-9.20	37	45.13	11.676	27.19	63.22	-332.07
Cadmium	mg/l	42	0.04	0.020	0.00	0.08	55.56	42	0.03	0.019	0.00	0.07	66.67
Chromium	mg/l	58	0.03	0.036	0.00	0.10	25.00	58	0.03	0.033	0.00	0.09	25.00
Copper	mg/l	63	0.04	0.029	0.01	0.09	75.00	63	0.04	0.035	0.00	0.10	75.00
Iron	mg/l	51	0.15	0.118	0.04	0.33	28.57	51	0.21	0.202	0.05	0.54	0.00
Potassium	mg/l	14	3.40	0.675	2.50	4.36	15.84	14	10.78	10.185	2.97	27.17	-166.83
Magnesium	mg/l	48	1.36	0.157	1.05	1.62	6.21	48	0.63	0.310	0.23	0.95	56.55
Manganese	mg/l	63	0.01	0.012	0.00	0.06	94.12	63	0.04	0.031	0.00	0.11	76.47
Sodium	mg/l	14	14.74	1.282	13.12	18.08	-2.93	14	15.90	1.869	13.87	18.80	-11.03
Nickel	mg/l	53	0.004	0.006	0.00	0.04	90.00	53	0.01	0.010	0.00	0.06	75.00
Zinc	mg/l	42	0.06	0.066	0.00	0.17	71.43	42	0.04	0.054	0.00	0.17	80.92
		2–day outflow (LC)–T7						*2–day outflow (LC)–T8*					
pH	–	85	7.5	0.70	6.3	10.1	na	36	10.6	0.99	8.5	12.5	na
Redox potential	mV	85	4.2	30.40	-116.1	51.0	na	36	-143.5	51.01	-238.1	-30.3	na
Turbidity	NTU	85	20.2	14.20	2.9	129.0	11.8	36	35.6	18.11	8.7	79.3	-55.5
Total suspended solids	mg/l	85	30.0	12.12	11.0	76.0	24.8	36	66.2	36.63	13.0	181.0	-65.9
Electrical conductivity	µS/cm	85	138.5	23.26	79.0	215.0	na	36	344.5	287.03	168.4	1534.0	na
Dissolved oxygen	mg/l	85	10.5	0.82	8.2	12.6	-1.0	36	10.1	0.73	6.5	10.8	2.9
Colour	Pa/Co	85	164.5	40.93	34.0	265.0	23.3	36	331.7	119.34	104.0	552.0	-54.6
Temperature	°C	85	16.0	4.59	5.3	21.8	na	36	16.3	4.24	6.3	21.3	na
Biochemical oxygen demand	mg/l	85	5.6	3.60	0.0	20.0	68.2	36	4.4	5.13	0.0	22.0	75.0
Chemical oxygen demand	mg/l	85	26.8	6.18	15.4	41.9	7.3	36	24.0	4.99	15.4	39.9	17.0
Ammonia–nitrogen	mg/l	85	0.09	0.05	0.0	0.3	55.0	36	0.1	0.04	0.1	0.2	50.0
Nitrate–nitrogen	mg/l	85	1.2	0.71	0.1	3.2	7.7	36	0.6	0.54	0.0	2.6	53.8
Ortho–phosphate–phosphorus	mg/l	85	7.0	3.89	3.0	18.8	16.7	36	3.9	1.25	2.2	7.1	53.6
Element													
Aluminium	mg/l	39	0.34	0.180	0.11	0.72	34.62	39	0.76	0.347	0.16	1.24	-46.15
Boron	mg/l	35	0.11	0.009	0.08	0.13	21.43	35	0.10	0.024	0.07	0.13	28.57
Calcium	mg/l	46	11.25	0.773	9.86	12.70	-6.74	37	70.99	33.166	21.66	109.98	-573.53
Cadmium	mg/l	42	0.05	0.031	0.00	0.11	44.44	42	0.04	0.030	0.00	0.10	55.56
Chromium	mg/l	58	0.04	0.049	0.00	0.12	0.00	58	0.05	0.039	0.00	0.12	-25.00
Copper	mg/l	63	0.06	0.049	0.02	0.15	62.50	63	0.05	0.043	0.01	0.13	68.75
Iron	mg/l	51	0.21	0.157	0.09	0.45	0.00	51	0.48	0.447	0.15	1.26	-128.57
Potassium	mg/l	14	3.87	0.364	3.35	4.50	4.21	14	12.77	15.139	2.73	36.71	-216.09
Magnesium	mg/l	48	1.35	0.133	0.99	1.58	6.90	48	0.70	0.336	0.28	1.15	51.72
Manganese	mg/l	63	0.08	0.056	0.00	0.18	52.94	63	0.08	0.069	0.00	0.20	52.94
Sodium	mg/l	14	13.82	1.175	12.14	15.57	3.49	14	15.35	3.197	12.32	20.34	-7.19
Nickel	mg/l	53	0.01	0.007	0.00	0.04	75.00	53	0.01	0.012	0.00	0.06	75.00
Zinc	mg/l	42	0.09	0.083	0.00	0.23	57.14	42	0.07	0.084	0.00	0.29	66.67

Note: LC, low pollutant concentrations; T5, treatment system with only *P. australis*; T6, treatment system with *P. australis* and ochre pellets; T7, treatment system without *P. australis* or ochre pellets; T8, treatment system with only ochre pellets; n, number of tested samples; SD, standard deviation; Min, minimum; Max, maximum; Rem, removal; NTU, nephelometric turbidity unit; na, not applicable.

Table 7.2. (Continued).

c) HC–SGW (7–day contact time)

		7–day outflow (HC)–T9						*7–day outflow (HC)–T10*					
Parameter	***Unit***	n	Mean	SD	Min	Max	Rem (%)	n	Mean	SD	Min	Max	Rem (%)
pH	–	83	7.3	0.82	5.9	8.8	na	34	9.8	1.34	7.3	12.3	na
Redox potential	mV	83	12.2	40.30	-57.6	77.5	na	34	-100.1	66.45	-217.4	22.3	na
Turbidity	NTU	83	154.8	86.08	9.8	430.0	18.1	34	178.8	98.79	23.8	356.0	5.3
Total suspended solids	mg/l	83	267.8	110.05	26.0	458.0	15.5	34	342.9	125.33	48.0	581.0	-8.2
Electrical conductivity	µS/cm	83	1137.4	471.09	475.0	3010.0	na	34	1191.1	343.72	733.0	1871.0	na
Dissolved oxygen	mg/l	83	8.8	0.89	6.5	11.0	16.2	34	8.3	1.03	6.4	10.7	21.0
Colour	Pa/Co	83	1448.1	647.98	106.0	2941.0	8.8	34	1593.5	761.50	226.0	2924.0	-0.4
Temperature	°C	83	16.8	4.03	7.2	22.5	na	34	18.0	4.14	7.3	22.6	na
Biochemical oxygen demand	mg/l	83	23.1	9.35	8.0	40.0	33.4	34	12.1	7.32	2.0	32.0	65.1
Chemical oxygen demand	mg/l	83	94.0	31.13	16.2	161.5	27.2	34	90.7	29.89	47.6	177.0	30.0
Ammonia–nitrogen	mg/l	83	0.5	0.23	0.2	1.2	-25.0	34	0.3	0.14	0.1	0.8	25.0
Nitrate–nitrogen	mg/l	83	10.7	7.92	0.9	37.9	-20.2	34	16.3	4.89	6.2	34.5	-83.1
Ortho–phosphate–phosphorus	mg/l	83	48.0	13.76	23.1	76.3	18.9	34	16.3	3.00	3.0	38.1	72.4
Element													
Aluminium	mg/l	54	2.33	1.321	0.21	5.12	-9.39	39	1.56	0.880	0.35	3.71	26.76
Boron	mg/l	26	0.55	0.211	0.33	0.92	3.50	23	0.44	0.202	0.26	0.91	22.81
Calcium	mg/l	52	42.49	4.386	31.32	48.65	-17.77	37	77.22	42.765	39.17	150.14	-114.02
Cadmium	mg/l	36	5.82	2.238	2.28	9.84	20.92	30	4.61	2.126	1.73	8.90	37.36
Chromium	mg/l	46	3.22	1.736	0.93	6.78	-0.63	40	2.86	1.328	1.24	5.16	10.63
Copper	mg/l	54	1.15	0.385	0.63	1.94	20.14	45	0.98	0.308	0.62	1.74	31.94
Iron	mg/l	42	5.45	1.657	3.34	9.40	14.98	36	5.03	1.475	2.81	6.86	21.53
Potassium	mg/l	8	44.90	2.827	41.21	48.07	25.37	8	56.58	19.919	36.93	77.55	5.95
Magnesium	mg/l	54	17.77	3.477	12.22	22.55	-3.55	42	12.84	6.124	5.36	22.80	25.17
Manganese	mg/l	54	0.35	0.249	0.09	0.82	64.29	45	0.46	0.212	0.19	0.77	53.06
Sodium	mg/l	8	55.09	11.391	42.72	66.71	12.11	8	55.85	12.850	42.23	74.21	10.90
Nickel	mg/l	50	0.10	0.091	0.00	0.23	-100.0	41	0.05	0.077	0.00	0.20	0.00
Zinc	mg/l	32	3.12	0.872	1.76	4.60	26.59	29	2.78	0.859	1.62	4.27	34.59
		7–day outflow (HC)–T11						*7–day outflow (HC)–T12*					
pH	–	83	7.7	1.21	5.9	9.9	na	34	9.8	1.54	7.1	12.3	na
Redox potential	mV	83	-4.4	59.67	-108.3	78.1	na	34	-95.5	88.21	-216.7	157.8	na
Turbidity	NTU	83	185.7	49.24	65.1	281.0	1.7	34	245.8	96.29	60.0	497.0	-30.1
Total suspended solids	mg/l	83	302.6	61.44	147.0	434.0	4.5	34	423.4	114.04	120.0	692.0	-33.6
Electrical conductivity	µS/cm	83	1003.0	306.88	492.0	2460.0	na	34	1107.1	299.47	734.0	1814.0	na
Dissolved oxygen	mg/l	83	10.5	0.91	7.9	12.0	0.0	34	9.8	1.19	5.4	11.7	6.7
Colour	Pa/Co	83	1644.8	489.96	718.0	2889.0	-3.6	34	2040.5	757.57	688.0	3282.0	-28.5
Temperature	°C	83	16.6	3.87	7.6	22.1	na	34	17.7	4.20	7.4	22.3	na
Biochemical oxygen demand	mg/l	83	16.6	7.07	4.0	38.0	52.2	34	8.3	4.23	0.0	18.0	76.1
Chemical oxygen demand	mg/l	83	100.8	27.65	11.6	159.5	22.0	34	103.1	16.10	75.6	135.0	20.2
Ammonia–nitrogen	mg/l	83	0.3	0.13	0.0	0.8	25.0	34	0.3	0.11	0.1	0.5	25.0
Nitrate–nitrogen	mg/l	83	8.5	8.42	0.4	34.5	4.5	34	15.0	8.59	3.7	38.7	-68.5
Ortho–phosphate–phosphorus	mg/l	83	43.0	13.78	20.25	79.4	27.2	34	17.3	5.63	3.8	32.8	70.7
Element													
Aluminium	mg/l	54	2.98	1.218	1.61	6.14	-39.91	24	3.61	2.306	0.87	6.67	-69.48
Boron	mg/l	26	0.54	0.160	0.34	0.77	5.26	20	0.39	0.078	0.30	0.51	31.58
Calcium	mg/l	52	37.39	4.030	30.58	45.66	-3.63	22	145.67	92.506	40.36	243.66	-303.74
Cadmium	mg/l	36	6.40	1.984	3.86	9.72	13.59	24	6.87	2.628	3.33	10.27	6.66
Chromium	mg/l	46	4.76	1.215	2.83	6.68	-48.75	34	4.75	2.021	2.57	6.89	-48.44
Copper	mg/l	54	1.30	0.301	0.80	1.76	9.72	36	1.47	0.247	1.14	1.80	-2.08
Iron	mg/l	42	7.02	1.801	3.58	9.36	-9.52	30	8.69	2.012	6.48	10.99	-35.57
Potassium	mg/l	8	45.77	5.160	39.87	51.00	23.92	8	59.62	20.132	39.79	79.86	0.90
Magnesium	mg/l	54	16.24	1.971	11.76	18.35	5.36	30	12.97	3.785	7.71	17.80	24.42
Manganese	mg/l	54	1.01	0.223	0.75	1.38	-3.06	36	0.86	0.457	0.33	1.42	12.24
Sodium	mg/l	8	55.22	11.852	41.86	67.68	11.90	8	55.59	12.232	42.05	68.09	11.31
Nickel	mg/l	50	0.09	0.081	0.00	0.20	-80.00	32	0.04	0.033	0.00	0.10	20.00
Zinc	mg/l	32	3.90	0.972	1.90	5.10	8.02	26	4.32	0.787	3.01	5.69	-1.65

Note: HC, high pollutant concentrations; T9, treatment system with only *P. australis*; T10, treatment system with *P. australis* and ochre pellets; T11, treatment system without *P. australis* or ochre pellets; T12, treatment system with ochre pellets only; n, number of tested samples; SD, standard deviation; Min, minimum; Max, maximum; Rem, removal; NTU, nephelometric turbidity unit; na, not applicable.

Table 7.2. (Continued).

d) LC–SGW (7–day contact time)

		7–day outflow (LC)–T13						*7–day outflow (LC)–T14*					
Parameter	***Unit***	n	Mean	SD	Min	Max	Rem (%)	n	Mean	SD	Min	Max	Rem (%)
pH	–	83	6.9	0.61	6.2	9.0	na	34	10.3	1.33	7.7	12.3	na
Redox potential	mV	83	31.0	28.12	-75.5	68.9	na	34	-130.8	63.74	-217.6	-1.8	na
Turbidity	NTU	83	18.9	11.05	2.8	52.1	17.5	34	25.1	16.21	2.9	77.8	-9.6
Total suspended solids	mg/l	83	27.7	16.48	2.8	72.0	30.6	34	37.5	15.62	4.0	74.0	6.0
Electrical conductivity	µS/cm	83	161.4	42.91	103.7	324.0	na	34	306.8	118.32	166.2	774.0	na
Dissolved oxygen	mg/l	83	9.3	1.24	4.9	12.0	10.6	34	8.7	0.94	7.0	10.7	16.3
Colour	Pa/Co	83	159.1	56.83	48.0	391.0	25.8	34	250.6	120.15	47.0	647.0	-16.8
Temperature	°C	83	15.9	4.18	7.3	22.4	na	34	17.3	4.31	7.3	22.3	na
Biochemical oxygen demand	mg/l	83	13.4	5.63	2.0	30.0	23.9	34	5.5	6.00	0.0	28.0	68.8
Chemical oxygen demand	mg/l	83	31.3	11.95	7.9	77.5	-8.3	34	29.2	10.71	20.3	63.4	-1.0
Ammonia–nitrogen	mg/l	83	0.1	0.07	0.0	0.5	50.0	34	0.1	0.07	0.0	0.3	50.0
Nitrate–nitrogen	mg/l	83	1.3	0.77	0.0	4.8	0.0	34	0.7	0.77	0.0	3.7	46.2
Ortho–phosphate–phosphorus	mg/l	83	11.9	6.36	3.3	32.0	-41.7	34	3.0	1.77	1.5	8.5	64.3
Element													
Aluminium	mg/l	44	0.12	0.094	0.00	0.24	76.92	24	0.37	0.232	0.08	0.69	28.85
Boron	mg/l	24	0.13	0.069	0.08	0.29	7.14	20	0.08	0.005	0.07	0.09	42.86
Calcium	mg/l	52	11.44	0.944	10.01	13.12	-8.54	22	60.11	13.881	38.50	75.27	-470.30
Cadmium	mg/l	32	0.08	0.097	0.00	0.27	11.11	24	0.02	0.021	0.00	0.06	77.78
Chromium	mg/l	42	0.05	0.069	0.00	0.21	-25.00	34	0.04	0.031	0.00	0.07	0.00
Copper	mg/l	48	0.07	0.081	0.00	0.22	56.25	36	0.04	0.032	0.00	0.08	75.00
Iron	mg/l	38	0.14	0.080	0.04	0.28	33.33	30	0.39	0.218	0.16	0.65	-85.71
Potassium	mg/l	8	2.99	0.216	2.72	3.29	25.99	8	17.59	16.141	2.35	33.47	-335.40
Magnesium	mg/l	46	1.55	0.195	1.18	1.91	-6.90	30	0.84	0.224	0.53	1.08	42.07
Manganese	mg/l	48	0.05	0.077	0.00	0.20	70.59	36	0.04	0.033	0.00	0.09	76.47
Sodium	mg/l	8	13.91	1.648	12.10	15.47	2.86	8	15.42	3.280	12.09	18.59	-7.68
Nickel	mg/l	44	0.05	0.081	0.00	0.18	-25.00	32	0.00	0.012	0.00	0.06	100.00
Zinc	mg/l	28	0.11	0.094	0.00	0.32	47.62	24	0.06	0.050	0.00	0.14	71.43
		7–day outflow (LC)–T15						*7–day outflow (LC)–T16*					
pH	–	83	7.5	0.72	6.4	9.3	na	34	10.5	1.05	8.0	12.2	na
Redox potential	mV	83	1.8	33.00	-87.9	53.2	na	34	-131.3	72.36	-217.6	156.0	na
Turbidity	NTU	83	16.5	7.27	5.7	34.1	27.9	34	40.9	25.03	4.0	113.0	-78.6
Total suspended solids	mg/l	83	25.0	10.96	7.0	56.0	37.3	34	55.2	24.85	4.0	104.0	-38.3
Electrical conductivity	µS/cm	83	144.0	32.28	97.7	263.0	na	34	290.2	135.74	148.0	768.0	na
Dissolved oxygen	mg/l	83	11.0	1.11	8.1	14.3	-5.8	34	10.1	0.84	8.3	11.2	2.9
Colour	Pa/Co	83	152.6	41.05	51.0	258.0	28.9	34	283.8	115.21	48.0	544.0	-32.3
Temperature	°C	83	15.3	4.23	6.7	22.2	na	34	17.0	4.15	7.9	22.1	na
Biochemical oxygen demand	mg/l	83	6.7	4.85	0.0	22.0	61.9	34	5.4	3.95	0.0	20.0	69.3
Chemical oxygen demand	mg/l	83	17.2	6.95	6.0	36.7	40.5	34	19.9	7.28	3.9	32.2	31.1
Ammonia–nitrogen	mg/l	83	0.1	0.04	0.0	0.3	50.0	34	0.1	0.15	0.0	0.8	50.0
Nitrate–nitrogen	mg/l	83	1.0	0.64	0.0	4.0	23.1	34	0.3	0.28	0.0	1.1	76.9
Ortho–phosphate–phosphorus	mg/l	83	8.5	4.03	2.6	19.6	-10.0	34	3.7	1.29	1.2	6.6	56.0
Element													
Aluminium	mg/l	44	0.36	0.189	0.09	0.75	30.77	24	0.73	0.420	0.20	1.40	-40.38
Boron	mg/l	24	0.12	0.064	0.08	0.26	14.29	20	0.08	0.006	0.07	0.09	42.86
Calcium	mg/l	52	10.74	0.739	9.44	12.12	-1.90	22	65.46	37.361	23.48	104.98	-521.06
Cadmium	mg/l	32	0.09	0.083	0.00	0.21	0.00	24	0.05	0.046	0.00	0.11	44.44
Chromium	mg/l	42	0.07	0.074	0.00	0.21	-75.00	34	0.06	0.054	0.00	0.12	-50.00
Copper	mg/l	48	0.10	0.091	0.00	0.26	37.50	36	0.06	0.057	0.00	0.13	62.50
Iron	mg/l	38	0.20	0.100	0.07	0.30	4.76	30	0.93	0.759	0.15	1.91	-342.86
Potassium	mg/l	8	3.62	0.438	3.07	4.22	10.40	8	20.16	19.003	2.26	38.75	-399.01
Magnesium	mg/l	46	1.38	0.161	1.03	1.64	4.83	30	0.78	0.330	0.36	1.16	46.21
Manganese	mg/l	48	0.06	0.074	0.00	0.21	64.71	36	0.10	0.094	0.00	0.21	41.18
Sodium	mg/l	8	13.15	1.199	11.83	14.36	8.17	8	15.69	5.272	10.55	21.03	-9.57
Nickel	mg/l	44	0.05	0.080	0.00	0.18	-25.00	32	0.01	0.010	0.00	0.05	75.00
Zinc	mg/l	28	0.13	0.068	0.01	0.25	38.10	24	0.11	0.089	0.00	0.26	47.62

Note: LC, low pollutant concentrations; T13, treatment system with only *P. australis*; T14, treatment system with *P. australis* and ochre pellets; T15, treatment system without *P. australis* or ochre pellets; T16, treatment system with only ochre pellets; n, number of tested samples; SD, standard deviation; Min, minimum; Max, maximum; Rem, removal; NTU, nephelometric turbidity unit; na, not applicable.

Table 7.2. (Continued).

e) outflow of control wetlands

		2–day outflow (TW)–C1					*2–day outflow (TW)–C2*				
Parameter	***Unit***	n	Mean	SD	Min	Max	n	Mean	SD	Min	Max
pH	–	85	6.7	0.39	6.0	7.7	85	7.4	0.60	6.2	9.3
Redox potential	mV	85	42.2	16.50	-7.7	70.8	85	9.6	28.10	-72.2	78.4
Turbidity	NTU	85	9.3	6.61	2.0	39.3	85	4.2	4.37	1.2	32.5
Total suspended solids	mg/l	85	14.3	8.16	0.0	35.0	85	3.9	2.93	0.0	15.0
Electrical conductivity	µS/cm	85	84.4	12.15	57.2	117.7	85	81.5	9.94	57.2	116.2
Dissolved oxygen	mg/l	85	9.0	0.87	7.2	11.8	85	10.4	0.70	8.9	12.0
Colour	Pa/Co	85	44.3	30.56	6.0	168.0	85	8.6	7.66	0.0	34.0
Temperature	°C	85	16.5	3.76	5.9	22.4	85	16.8	4.04	6.0	22.8
Biochemical oxygen demand	mg/l	85	7.3	3.45	0.0	18.0	85	5.4	4.03	0.0	18.0
Chemical oxygen demand	mg/l	85	15.9	7.74	4.9	42.8	85	6.3	2.84	1.0	14.3
Ammonia–nitrogen	mg/l	85	0.1	0.12	0.0	0.6	85	0.1	0.14	0.0	0.6
Nitrate–nitrogen	mg/l	85	1.1	0.75	0.1	3.8	85	0.8	0.53	0.0	3.6
Ortho–phosphate–phosphorus	mg/l	85	2.8	1.82	0.9	10.6	85	2.4	0.63	0.9	4.3
Element											
Aluminium	mg/l	24	0.01	0.006	0.00	0.02	24	0.01	0.007	0.00	0.02
Boron	mg/l	20	0.02	0.018	0.00	0.05	20	0.03	0.009	0.01	0.05
Calcium	mg/l	28	9.96	0.549	9.06	10.90	28	9.78	0.552	8.90	10.67
Cadmium	mg/l	24	0.01	0.006	0.00	0.02	24	0.00	0.006	0.00	0.02
Chromium	mg/l	34	0.00	0.005	0.00	0.01	34	0.00	0.005	0.00	0.01
Copper	mg/l	36	0.01	0.006	0.00	0.02	36	0.01	0.008	0.00	0.03
Iron	mg/l	30	0.02	0.007	0.00	0.04	30	0.02	0.009	0.00	0.04
Potassium	mg/l	8	0.35	0.049	0.27	0.42	8	0.69	0.261	0.40	0.98
Magnesium	mg/l	30	1.10	0.123	0.77	1.27	30	1.10	0.138	0.78	1.30
Manganese	mg/l	36	0.01	0.010	0.00	0.04	36	0.00	0.009	0.00	0.03
Sodium	mg/l	8	6.62	0.721	5.82	7.31	8	6.69	0.869	5.78	7.58
Nickel	mg/l	32	0.01	0.023	0.00	0.09	32	0.01	0.023	0.00	0.10
Zinc	mg/l	26	0.03	0.009	0.00	0.03	26	0.02	0.010	0.00	0.04
		7–day outflow (TW)–C3					*7–day outflow (TW)–C4*				
pH	–	83	6.6	0.39	6.0	7.3	83	7.1	0.52	5.9	8.9
Redox potential	mV	83	44.1	17.06	6.1	76.4	83	25.1	24.68	-60.9	89.8
Turbidity	NTU	83	12.7	12.56	1.5	69.9	83	3.7	3.47	1.3	30.1
Total suspended solids	mg/l	83	17.8	13.69	0.0	67.0	83	4.3	5.79	0.0	38.0
Electrical conductivity	µS/cm	83	92.9	27.28	60.7	167.2	83	87.1	20.83	58.4	163.2
Dissolved oxygen	mg/l	83	8.9	1.09	5.7	11.2	83	10.8	1.07	8.4	13.2
Colour	Pa/Co	83	56.1	31.45	11.0	143.0	83	12.7	9.73	0.0	42.0
Temperature	°C	83	15.1	4.20	6.2	22.2	83	15.5	4.17	7.2	22.0
Biochemical oxygen demand	mg/l	83	9.1	5.05	0.0	22.0	83	6.7	4.65	0.0	24.0
Chemical oxygen demand	mg/l	83	17.6	6.74	6.3	35.2	83	7.0	2.48	1.6	17.3
Ammonia–nitrogen	mg/l	83	0.1	0.04	0.0	0.2	83	0.1	0.05	0.0	0.2
Nitrate–nitrogen	mg/l	83	0.9	0.42	0.0	2.8	83	0.8	0.54	0.0	3.0
Ortho–phosphate–phosphorus	mg/l	83	3.4	1.47	0.4	7.4	83	2.4	0.86	0.5	4.6
Element											
Aluminium	mg/l	39	0.08	0.092	0.00	0.20	39	0.09	0.101	0.00	0.22
Boron	mg/l	23	0.05	0.061	0.00	0.19	23	0.05	0.059	0.00	0.19
Calcium	mg/l	49	9.67	0.591	8.41	10.70	49	9.51	0.476	8.40	10.40
Cadmium	mg/l	30	0.04	0.071	0.00	0.19	30	0.05	0.071	0.00	0.20
Chromium	mg/l	40	0.03	0.063	0.00	0.19	40	0.03	0.063	0.00	0.19
Copper	mg/l	45	0.04	0.073	0.00	0.19	45	0.05	0.078	0.00	0.20
Iron	mg/l	36	0.05	0.069	0.00	0.22	36	0.05	0.066	0.00	0.20
Potassium	mg/l	8	0.50	0.492	0.03	1.01	8	0.52	0.127	0.39	0.70
Magnesium	mg/l	42	1.20	0.119	0.88	1.40	42	1.16	0.120	0.83	1.37
Manganese	mg/l	45	0.04	0.070	0.00	0.19	45	0.04	0.069	0.00	0.18
Sodium	mg/l	8	6.80	0.085	6.69	6.90	8	6.35	0.105	6.24	6.56
Nickel	mg/l	41	0.04	0.075	0.00	0.18	41	0.04	0.075	0.00	0.18
Zinc	mg/l	29	0.04	0.070	0.00	0.24	29	0.04	0.061	0.00	0.22

Note: TW, tap water; C1 and C3, treatment system with only *P. australis*; C2 and C4, treatment system with only tap water (TW); n, number of tested samples; SD, standard deviation; Min, minimum; Max, maximum; Rem, removal; NTU, nephelometric turbidity unit; na, not applicable.

7.2.1. Temperature, pH, Redox Potential and Electrical Conductivity

The overall mean values of temperature for inflow were around 16.9 °C and 17.7 °C for HC– and LC–SGW, respectively. The statistical analysis showed that temperature did not significantly change ($p > 0.05$) when comparing the inflow SGW with outflows from all treatment systems of different design set–ups (Table 7.3). In this study, the pH had a mean value of 8.4±1.61 for inflow HC–SGW, while it was close to neutrality (6.9±0.48) for the inflow LC–SGW. It was clearly noted that pH correlated negatively significantly ($r = - 0.967$; $p < 0.001$) with Eh value in all treatment systems, which is a known fact in aqueous ecosystems (Bezbaruah and Zhang, 2004).

In summary, the cement–ochre pellets utilised in current investigation, had pH values between 9.63 and 12.53, with associated redox potentials between -248.5 and -98.8 mV. The average electrical conductivity range of those pellets was 1580–2300 µS/cm. Therefore, the presence of ochre pellets in the treatment systems always significantly increased pH and EC of the outflows, since there was a positive significant correlation ($r = 0.717$, $p = 0.030$) between Ca and pH. Calcium is released from those pellets to the SGW and subsequently increases the pH of the outflows (Heal et al., 2003). However, presence of *P. australis* in treatment SGW significantly decreased the pH value of the outflow compared with inflow (Table 7.3). The drop in pH could be due to production of carbon dioxide during rhizome respiration (Ijaz et al., 2015; Abed et al., 2017), and/or organic acids which are by–products of biodegradation of organic substances in water by microorganisms (Bezbaruah and Zhang, 2004, Iamchaturapatr et al., 2007).

Table 7.3. Significant values of the statistical analysis for the comparison between treated and untreated greywater samples; a) high concentration greywater (HC–SGW) at 2–day contact time, b) low concentration greywater (LC–SGW) at 2–day contact time, c) high concentration greywater (HC–SGW) at 7–day contact time, and d) low concentration greywater (LC–SGW) at 7–day contact time.

a) HC–SGW: comparisons at 2–day contact time (inflow & outflow)

Parameter	*Unit*	*Inflow & T1*			*Inflow & T2*			*Inflow & T3*			*Inflow & T4*		
		Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
pH	–	<0.001	M–W	<0.001	0.147	T–test	0.183	<0.001	M–W	0.027	0.149	T–test	0.313
Redox potential	mV	<0.001	M–W	<0.001	0.086	T–test	0.239	<0.001	M–W	0.004	0.081	T–test	0.390
Turbidity	NTU	0.273	T–test	0.121	0.001	M–W	0.317	0.118	T–test	0.676	<0.001	M–W	0.071
Total suspended solids	mg/l	0.113	T–test	0.179	<0.001	M–W	0.001	0.217	T–test	0.593	<0.001	M–W	0.603
Electrical conductivity	µS/cm	0.018	M–W	0.904	<0.001	M–W	0.001	0.012	M–W	0.376	<0.001	M–W	<0.001
Dissolved oxygen	mg/l	0.010	M–W	<0.001	0.003	M–W	<0.001	0.005	M–W	0.002	0.007	M–W	0.001
Colour	Pa/Co	0.734	T–test	0.313	<0.001	M–W	0.001	0.218	T–test	0.274	<0.001	M–W	0.032
Temperature	°C	<0.001	M–W	0.678	0.003	M–W	0.770	0.005	M–W	0.923	0.005	M–W	0.939
Biochemical oxygen demand	mg/l	<0.001	M–W	<0.001	0.001	M–W	<0.001	0.002	M–W	<0.001	0.002	M–W	<0.001
Chemical oxygen demand	mg/l	0.008	M–W	<0.001	0.060	T–test	0.001	0.005	M–W	<0.001	0.014	M–W	<0.001
Ammonia–nitrogen	mg/l	<0.001	M–W	0.269	<0.001	M–W	0.050	<0.001	M–W	0.860	<0.001	M–W	0.545
Nitrate–nitrogen	mg/l	0.006	M–W	<0.001	0.016	M–W	<0.001	0.005	M–W	0.325	<0.001	M–W	0.006
Ortho–phosphate–phosphorus	mg/l	0.330	T–test	0.002	0.002	M–W	<0.001	0.107	T–test	<0.001	<0.001	M–W	<0.001
Aluminium (Al)	mg/l	0.001	M–W	0.002	<0.001	M–W	0.083	0.009	M–W	0.171	<0.001	M–W	0.696
Boron (B)	mg/l	<0.001	M–W	0.006	<0.001	M–W	<0.001	0.002	M–W	0.171	0.004	M–W	<0.001
Calcium (Ca)	mg/l	0.048	M–W	0.001	<0.001	M–W	<0.001	0.001	M–W	0.001	<0.001	M–W	<0.001
Cadmium (Cd)	mg/l	<0.001	M–W	0.007	<0.001	M–W	<0.001	<0.001	M–W	0.159	<0.001	M–W	0.128
Chromium (Cr)	mg/l	<0.001	M–W	<0.001	<0.001	M–W	0.005	0.004	M–W	0.003	<0.001	M–W	0.104
Copper (Cu)	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	0.378	<0.001	M–W	0.005
Iron (Fe)	mg/l	<0.001	M–W	0.001	<0.001	M–W	0.019	0.002	M–W	0.867	<0.001	M–W	0.140
Potassium (K)	mg/l	0.013	M–W	<0.001	0.002	M–W	0.064	0.004	M–W	0.014	0.003	M–W	0.064
Magnesium (Mg)	mg/l	<0.001	M–W	0.348	<0.001	M–W	<0.001	0.007	M–W	0.120	<0.001	M–W	<0.001
Manganese (Mn)	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	0.006	<0.001	M–W	<0.001
Sodium (Na)	mg/l	0.005	M–W	0.328	0.009	M–W	0.355	0.007	M–W	0.304	0.009	M–W	0.411
Nickel (Ni)	mg/l	<0.001	M–W	0.005	<0.001	M–W	<0.001	<0.001	M–W	0.169	<0.001	M–W	0.007
Zinc (Zn)	mg/l	<0.001	M–W	0.004	<0.001	M–W	<0.001	0.004	M–W	0.500	<0.001	M–W	0.208

Note: T1, treatment system with *P. australis* only; T2, treatment system with *P. australis* and ochre pellets; T3, treatment system without *P. australis* or ochre pellets; T4, treatment system with ochre pellets only; Shapiro–Wilk (check for normality), normally distributed data if $p > 0.05$ using T–test and non–normally distributed data if $p < 0.05$ using Mann–Whitney U–test; p value, significant difference if $p < 0.05$ and not significant if $p > 0.05$; M–W, Mann–Whitney U–test; NTU, nephelometric turbidity unit.

Table 7.3. (Continued).

b) LC–SGW: comparisons at 2–day contact time (inflow & outflow)

Parameter	***Unit***	***Inflow & T5***			***Inflow & T6***			***Inflow & T7***			***Inflow & T8***		
		Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
pH	–	0.009	M–W	0.821	<0.001	M–W	<0.001	0.132	T–test	<0.001	<0.001	M–W	<0.001
Redox potential	mV	<0.001	M–W	0.494	<0.001	M–W	<0.001	0.013	M–W	<0.001	<0.001	M–W	<0.001
Turbidity	NTU	<0.001	M–W	0.258	<0.001	M–W	0.016	<0.001	M–W	0.004	<0.001	M–W	<0.001
Total suspended solids	mg/l	<0.001	M–W	0.050	<0.001	M–W	0.009	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Electrical conductivity	µS/cm	<0.001	M–W	0.061	<0.001	M–W	<0.001	<0.001	M–W	0.001	<0.001	M–W	<0.001
Dissolved oxygen	mg/l	0.001	M–W	<0.001	0.012	M–W	<0.001	<0.001	M–W	0.846	<0.001	M–W	0.009
Colour	Pa/Co	0.963	T–test	0.005	<0.001	M–W	0.001	0.483	T–test	<0.001	0.003	M–W	0.032
Temperature	°C	0.002	M–W	0.432	0.003	M–W	0.215	0.015	M–W	0.053	0.009	M–W	0.096
Biochemical oxygen demand	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Chemical oxygen demand	mg/l	<0.001	M–W	0.060	<0.001	M–W	0.725	<0.001	M–W	0.811	<0.001	M–W	0.199
Ammonia–nitrogen	mg/l	<0.001	M–W	0.003	<0.001	M–W	0.166	<0.001	M–W	<0.001	<0.001	M–W	0.037
Nitrate–nitrogen	mg/l	<0.001	M–W	0.003	<0.001	M–W	<0.001	<0.001	M–W	0.687	<0.001	M–W	<0.001
Ortho–phosphate–phosphorus	mg/l	<0.001	M–W	0.319	<0.001	M–W	<0.001	<0.001	M–W	0.003	<0.001	M–W	<0.001
Aluminium (Al)	mg/l	<0.001	M–W	<0.001	<0.001	M–W	0.027	<0.001	M–W	0.192	<0.001	M–W	0.080
Boron (B)	mg/l	<0.001	M–W	0.009	<0.001	M–W	<0.001	<0.001	M–W	0.001	<0.001	M–W	0.001
Calcium (Ca)	mg/l	0.059	T–test	<0.001	<0.001	M–W	<0.001	0.387	T–test	<0.001	<0.001	M–W	<0.001
Cadmium (Cd)	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Chromium (Cr)	mg/l	<0.001	M–W	0.071	<0.001	M–W	0.470	<0.001	M–W	0.014	<0.001	M–W	0.005
Copper (Cu)	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Iron (Fe)	mg/l	<0.001	M–W	<0.001	<0.001	M–W	0.024	<0.001	M–W	0.001	<0.001	M–W	0.051
Potassium (K)	mg/l	0.122	T–test	0.008	0.167	T–test	0.028	0.125	T–test	0.309	0.536	T–test	0.050
Magnesium (Mg)	mg/l	<0.001	M–W	0.021	<0.001	M–W	<0.001	<0.001	M–W	0.065	<0.001	M–W	<0.001
Manganese (Mn)	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Sodium (Na)	mg/l	0.186	T–test	0.476	0.007	M–W	0.090	0.010	M–W	0.382	0.043	M–W	0.681
Nickel (Ni)	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	0.410
Zinc (Zn)	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001

Note: T9, treatment system with *P. australis* only; T10, treatment system with *P. australis* and ochre pellets; T11, treatment system without *P. australis* or ochre pellets; T12, treatment system with ochre pellets only; Shapiro–Wilk (check for normality), normally distributed data if $p > 0.05$ using T–test and non–normal distributed data if $p < 0.05$ using Mann–Whitney U–test; p value, significant difference if $p < 0.05$ and not significant if $p > 0.05$; M–W, Mann–Whitney U–test; NTU, nephelometric turbidity unit.

Table 7.3. (Continued).

c) HC–SGW: comparisons at 7–day contact time (inflow & outflow)

Parameter	***Unit***	***Inflow & T9***			***Inflow & T10***			***Inflow & T11***			***Inflow & T12***		
		Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
pH	–	<0.001	M–W	<0.001	0.002	M–W	<0.001	<0.001	M–W	0.004	0.021	M–W	<0.001
Redox potential	mV	<0.001	M–W	<0.001	0.006	M–W	<0.001	<0.001	M–W	0.004	0.099	T–test	<0.001
Turbidity	NTU	0.003	M–W	0.008	0.086	T–test	0.573	0.471	T–test	0.667	<0.001	M–W	<0.001
Total suspended solids	mg/l	0.018	M–W	0.014	0.009	M–W	0.006	0.318	T–test	0.127	<0.001	M–W	<0.001
Electrical conductivity	µS/cm	0.216	T–test	0.009	<0.001	M–W	0.002	0.042	M–W	0.320	<0.001	M–W	0.149
Dissolved oxygen	mg/l	0.001	M–W	<0.001	0.001	M–W	<0.001	0.004	M–W	0.301	0.001	M–W	0.003
Colour	Pa/Co	0.313	T–test	0.094	0.615	T–test	0.967	0.285	T–test	0.405	0.001	M–W	0.001
Temperature	°C	0.002	M–W	0.730	0.002	M–W	0.371	0.004	M–W	0.585	0.003	M–W	0.502
Biochemical oxygen demand	mg/l	<0.001	M–W	<0.001	0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Chemical oxygen demand	mg/l	0.040	M–W	<0.001	0.022	M–W	<0.001	0.002	M–W	<0.001	0.001	M–W	<0.001
Ammonia–nitrogen	mg/l	<0.001	M–W	<0.001	<0.001	M–W	0.001	<0.001	M–W	0.091	<0.001	M–W	0.009
Nitrate–nitrogen	mg/l	0.002	M–W	0.266	0.006	M–W	<0.001	<0.001	M–W	0.204	<0.001	M–W	<0.001
Ortho–phosphate–phosphorus	mg/l	0.146	T–test	<0.001	<0.001	M–W	<0.001	0.195	T–test	<0.001	<0.001	M–W	<0.001
Aluminium (Al)	mg/l	0.002	M–W	0.302	0.045	M–W	0.007	0.156	T–test	<0.001	<0.001	M–W	0.104
Boron (B)	mg/l	<0.001	M–W	0.099	0.002	M–W	<0.001	0.016	M–W	0.593	0.018	M–W	<0.001
Calcium (Ca)	mg/l	<0.001	M–W	0.001	0.032	M–W	<0.001	0.003	M–W	0.068	<0.001	M–W	<0.001
Cadmium (Cd)	mg/l	<0.001	M–W	0.075	<0.001	M–W	0.001	<0.001	M–W	0.340	<0.001	M–W	0.119
Chromium (Cr)	mg/l	0.001	M–W	0.688	0.001	M–W	0.230	<0.001	M–W	<0.001	<0.001	M–W	0.119
Copper (Cu)	mg/l	0.001	M–W	0.011	0.001	M–W	<0.001	0.001	M–W	0.177	0.001	M–W	0.436
Iron (Fe)	mg/l	0.002	M–W	0.064	0.001	M–W	0.009	0.003	M–W	0.193	<0.001	M–W	<0.001
Potassium (K)	mg/l	0.002	M–W	<0.001	0.006	M–W	1.000	0.002	M–W	<0.001	0.011	M–W	1.000
Magnesium (Mg)	mg/l	0.001	M–W	0.169	<0.001	M–W	0.001	0.024	M–W	0.277	<0.001	M–W	<0.001
Manganese (Mn)	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	0.023	<0.001	M–W	0.595
Sodium (Na)	mg/l	0.004	M–W	0.143	0.002	M–W	0.165	0.005	M–W	0.165	0.004	M–W	0.165
Nickel (Ni)	mg/l	<0.001	M–W	0.012	<0.001	M–W	0.182	<0.001	M–W	0.016	<0.001	M–W	0.574
Zinc (Zn)	mg/l	<0.001	M–W	0.006	<0.001	M–W	<0.001	0.001	M–W	0.503	0.001	M–W	0.538

Note: T9, treatment system with *P. australis* only; T10, treatment system with *P. australis* and ochre pellets; T11, treatment system without *P. australis* or ochre pellets; T12, treatment system with ochre pellets only; Shapiro–Wilk (check for normality), normally distributed data if $p > 0.05$ using T–test and non–normal distributed data if $p < 0.05$ using Mann–Whitney U–test; p value, significant difference if $p < 0.05$ and not significant if $p > 0.05$; M–W, Mann–Whitney U–test; NTU, nephelometric turbidity unit.

Table 7.3. (Continued).

d) LC–SGW: comparisons at 7–day contact time (inflow & outflow)

Parameter	***Unit***	***Inflow & T13***			***Inflow & T14***			***Inflow & T15***			***Inflow & T16***		
		Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
pH	–	<0.001	M–W	0.547	<0.001	M–W	<0.001	0.005	M–W	<0.001	<0.001	M–W	<0.001
Redox potential	mV	0.005	M–W	0.140	0.005	M–W	<0.001	0.005	M–W	<0.001	0.005	M–W	<0.001
Turbidity	NTU	0.105	T–test	0.007	0.005	M–W	0.811	0.008	M–W	<0.001	0.005	M–W	<0.001
Total suspended solids	mg/l	0.002	M–W	<0.001	<0.001	M–W	0.847	<0.001	M–W	<0.001	0.002	M–W	<0.001
Electrical conductivity	µS/cm	<0.001	M–W	0.765	<0.001	M–W	<0.001	<0.001	M–W	0.013	<0.001	M–W	<0.001
Dissolved oxygen	mg/l	0.003	M–W	<0.001	0.001	M–W	<0.001	<0.001	M–W	0.005	<0.001	M–W	0.068
Colour	Pa/Co	0.173	T–test	<0.001	<0.001	M–W	0.230	0.245	T–test	<0.001	0.100	T–test	0.002
Temperature	°C	0.002	M–W	0.009	0.001	M–W	0.783	0.002	M–W	0.001	0.002	M–W	0.392
Biochemical oxygen demand	mg/l	<0.001	M–W	0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Chemical oxygen demand	mg/l	<0.001	M–W	0.029	<0.001	M–W	0.506	<0.001	M–W	<0.001	<0.001	M–W	0.001
Ammonia–nitrogen	mg/l	<0.001	M–W	0.001	<0.001	M–W	0.169	<0.001	M–W	<0.001	<0.001	M–W	0.019
Nitrate–nitrogen	mg/l	<0.001	M–W	0.408	<0.001	M–W	0.001	<0.001	M–W	0.201	<0.001	M–W	<0.001
Ortho–phosphate–phosphorus	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	0.606	<0.001	M–W	<0.001
Aluminium (Al)	mg/l	<0.001	M–W	<0.001	<0.001	M–W	0.378	<0.001	M–W	0.321	<0.001	M–W	0.025
Boron (B)	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Calcium (Ca)	mg/l	0.001	M–W	<0.001	<0.001	M–W	<0.001	0.079	T–test	0.201	<0.001	M–W	<0.001
Cadmium (Cd)	mg/l	<0.001	M–W	0.001	<0.001	M–W	<0.001	<0.001	M–W	0.290	<0.001	M–W	0.050
Chromium (Cr)	mg/l	<0.001	M–W	0.717	<0.001	M–W	0.945	<0.001	M–W	0.360	<0.001	M–W	0.738
Copper (Cu)	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Iron (Fe)	mg/l	<0.001	M–W	0.007	<0.001	M–W	<0.001	<0.001	M–W	0.444	<0.001	M–W	<0.001
Potassium (K)	mg/l	0.151	T–test	<0.001	<0.001	M–W	1.000	0.334	T–test	0.054	<0.001	M–W	1.000
Magnesium (Mg)	mg/l	<0.001	M–W	0.006	<0.001	M–W	<0.001	<0.001	M–W	0.159	<0.001	M–W	<0.001
Manganese (Mn)	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Sodium (Na)	mg/l	0.001	M–W	0.316	0.011	M–W	0.396	0.012	M–W	0.165	0.028	M–W	1.000
Nickel (Ni)	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Zinc (Zn)	mg/l	<0.001	M–W	0.045	<0.001	M–W	<0.001	<0.001	M–W	0.328	<0.001	M–W	0.010

Note: T13, treatment system with *P. australis* only; T14, treatment system with *P. australis* and ochre pellets; T15, treatment system without *P. australis* or ochre pellets; T16, treatment system with ochre pellets only; Shapiro–Wilk (check for normality), normally distributed data if $p > 0.05$ using T–test and non–normally distributed data if $p < 0.05$ using Mann–Whitney U–test; p value, significant difference if $p < 0.05$ and not significant if $p > 0.05$; M–W, Mann–Whitney U–test; NTU, nephelometric turbidity unit.

The statistical analysis showed that *P. australis* unable to cope the effect of ochre pellets in terms of pH and EC, when they presented together. Since, there were no significant effect on pH values when *P. australis* appeared in combination with ochre pellets in wetlands T2 (HC–SGW, 2–day), T6 (LC–SGW, 2–day), T10 (HC–SGW, 7–day), and T14 (LC–SGW,7–day), treating both types of SGWs at both contact times compared to those values of outflows from systems treating SGWs with only ochre pellets such as T4 (HC–SGW, 2–day), T8 (LC–SGW, 2–day), T12(HC–SGW, 7–day), and T16 (LC–SGW, 7–day), respectively (Table 7.4). Furthermore, increasing contact time of treatment of SGW in wetland systems using a combination of *P. australis* and ochre pellets significantly raised the pH value, due to the overwhelming effect of ochre pellets, in comparing T2 and T6 with T10 and T14, respectively (Table 7.4).

7.2.2. Turbidity, Total Suspended Solids and Colour

In this study, outflows of all systems were agitated before sampling to focus on removal mechanisms other than sedimentation, such as the role of *P. australis*, ochre pellets or their combination.

Due to the relatively high level of pH associated with using ochre pellets in contact with water in systems T4, T8, T12 and T16, it could be that the precipitation of dissolved solids in SGW and constituent pigment of ochre led to significant increasing ($p < 0.05$) in turbidity, TSS and colour values of outflows compared to the inflow (Heal et al., 2005; Dobbie et al., 2009). The statistical analysis showed that pH correlated positively significantly with turbidity, TSS and colour as ($r = 0.700$, $p = 0.036$), ($r = 0.950$, $p < 0.001$) and ($r = 0.783$, $p = 0.013$), respectively. On other hand, presence of *P. australis* in floating systems treating SGW (T1, T5, T9 and T13) resulted in a significant drop in turbidity, TSS and colour (Table 7.4).

The rhizome systems of the *P. australis* and the biofilm developed on them and on the vessel walls led to improve the processes of filtration and biodegradation supporting the removal of TSS and turbidity with subsequent colour removal (Cheng et al., 2009; Abed et al., 2017). The correlations between turbidity and TSS, Turbidity and colour, and TSS and colour were positively significant ($r = 0.783$, $p = 0.013$; $r = 0.767$, $p = 0.016$; and $r = 0.767$, $p = 0.016$, respectively). However, on some occasions, sloughing off of heavy biofilms from rhizomes surfaces with attached trapped solids, in systems of floating macrophytes, could be causing an increase in TSS concentrations, as in comparison of outflow of T2 to T4 (Headley and Tanner, 2012), as shown in Table 7.4.

Therefore, presence of *P. australis* could mitigate the effect of ochre pellets when combined together in treatment of both types of SGWs (as in treatment systems T2, T6, T10 and T14) by significant reduction of turbidity, TSS and colour (Table 7.4).

Furthermore, the only significant effect of increasing contact time of the treatment of SGW was a decrease in the TSS and colour in systems with a combination of ochre pellets and *P. australis* (T10 and T14), compared with systems of 2–day contact time (T2 and T6, respectively). There were no significant differences in the removals of turbidity, TSS and colour in systems using a combination of ochre pellets with *P. australis*, when comparing treatment of LC–SGW (T6 and T14) with HC–SGW (T2 and T10, respectively), at both contact times (Table 7.4).

Table 7.4. Significant values of the statistical analysis for treated greywater samples to investigate *(a)* the effects of cement–ochre pellets on greywater remediation of the floating treatment wetlands (T), *(b)* the effects of *P. australis* on greywater remediation of treatment systems (T) with cement–ochre pellets, and *(c)* the effects of contact time and pollutant loads on greywater remediation in treatment systems (T) containing floating *P. australis* and ochre pellets.

(a) Effects of cement–ochre pellets on greywater remediation in floating *P. australis* treatment systems (T)

		2–day contact time						***7–day contact time***					
		HC–SGW (T1 & T2)			**LC–SGW (T5 & T6)**			**HC–SGW (T9 & T10)**			**LC–SGW (T13 & T14)**		
Parameter	***Unit***	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
pH	–	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Redox potential	mV	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Turbidity	NTU	0.072	T–test	0.009	<0.001	M–W	0.011	0.024	M–W	0.310	<0.001	M–W	0.061
Total suspended solids	mg/l	<0.001	M–W	<0.001	<0.001	M–W	0.001	0.006	M–W	0.001	0.008	M–W	0.003
Electrical conductivity	µS/cm	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	0.157	<0.001	M–W	<0.001
Dissolved oxygen	mg/l	0.014	M–W	0.939	0.218	T–test	0.008	0.778	T–test	0.004	0.426	T–test	0.007
Colour	Pa/Co	<0.001	M–W	<0.001	<0.001	M–W	<0.001	0.218	T–test	0.298	<0.001	M–W	<0.001
Temperature	°C	<0.001	M–W	0.968	<0.001	M–W	0.341	<0.001	M–W	0.055	<0.001	M–W	0.053
Biochemical oxygen demand	mg/l	0.105	T–test	<0.001	<0.001	M–W	<0.001	0.001	M–W	<0.001	0.013	M–W	<0.001
Chemical oxygen demand	mg/l	0.079	T–test	0.032	<0.001	M–W	0.135	0.048	M–W	0.442	<0.001	M–W	0.181
Ammonia–nitrogen	mg/l	<0.001	M–W	0.230	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	0.090
Nitrate–nitrogen	mg/l	0.071	T–test	0.880	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Ortho–phosphate–phosphorus	mg/l	0.001	M–W	<0.001	<0.001	M–W	<0.001	0.005	M–W	<0.001	<0.001	M–W	<0.001
Aluminium	mg/l	<0.001	M–W	0.017	<0.001	M–W	<0.001	0.001	M–W	0.004	<0.001	M–W	<0.001
Boron	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	0.012	<0.001	M–W	<0.001
Calcium	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Cadmium	mg/l	<0.001	M–W	0.809	<0.001	M–W	0.008	0.001	M–W	0.031	<0.001	M–W	0.263
Chromium	mg/l	<0.001	M–W	0.143	<0.001	M–W	0.417	<0.001	M–W	0.622	<0.001	M–W	0.925
Copper	mg/l	<0.001	M–W	0.229	<0.001	M–W	0.821	<0.001	M–W	0.029	<0.001	M–W	0.538
Iron	mg/l	<0.001	M–W	0.039	<0.001	M–W	0.100	<0.001	M–W	0.405	<0.001	M–W	<0.001
Potassium	mg/l	0.498	T–test	0.686	0.018	M–W	0.002	0.001	M–W	1.000	<0.001	M–W	1.000
Magnesium	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Manganese	mg/l	<0.001	M–W	0.117	<0.001	M–W	<0.001	<0.001	M–W	0.049	<0.001	M–W	0.269
Sodium	mg/l	0.322	T–test	0.687	0.272	T–test	0.067	0.005	M–W	0.834	0.003	M–W	0.462
Nickel	mg/l	<0.001	M–W	0.073	<0.001	M–W	0.050	<0.001	M–W	0.002	<0.001	M–W	0.365
Zinc	mg/l	<0.001	M–W	0.325	<0.001	M–W	0.240	<0.001	M–W	0.109	<0.001	M–W	0.010

Note: HC, high concentration; LC, low concentration; SGW, synthetic greywater; T1, T5, T9, and T13, treatment systems with only floating *P. australis*; T2, T6, T10, and T14, treatment systems with floating *P. australis* and ochre pellets; Shapiro–Wilk (check for normality), normally distributed data if $p > 0.05$ using T–test and non–normally distributed data if $p < 0.05$ using Mann–Whitney U–test; p value, significantly different, if $p < 0.05$ and not significantly different if $p > 0.05$; M–W, Mann–Whitney U–test; NTU, nephelometric turbidity unit.

Table 7.4. (Continued).

(b) Effects of *P. australis* on greywater remediation in treatment systems with ochre pellets (T)

		2–day contact time						***7–day contact time***					
		HC–SGW (T2 & T4)			**LC–SGW (T6 & T8)**			**HC–SGW (T10 & T12)**			**LC–SGW (T14 & T16)**		
Parameter	***Unit***	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
pH	–	<0.001	M–W	0.517	0.122	T–test	0.672	0.008	M–W	0.528	<0.001	M–W	0.440
Redox potential	mV	<0.001	M–W	0.517	0.109	T–test	0.626	0.006	M–W	0.736	<0.001	M–W	0.704
Turbidity	NTU	<0.001	M–W	0.285	<0.001	M–W	0.292	0.161	T–test	0.006	<0.001	M–W	0.001
Total suspended solids	mg/l	0.001	M–W	0.037	<0.001	M–W	0.186	0.029	M–W	0.011	0.182	T–test	0.001
Electrical conductivity	µS/cm	<0.001	M–W	0.879	<0.001	M–W	0.199	<0.001	M–W	0.272	<0.001	M–W	0.177
Dissolved oxygen	mg/l	0.005	M–W	<0.001	<0.001	M–W	<0.001	0.563	T–test	<0.001	0.042	M–W	<0.001
Colour	Pa/Co	0.046	M–W	0.392	0.015	M–W	0.362	0.174	T–test	0.018	0.085	T–test	0.249
Temperature	°C	<0.001	M–W	0.543	<0.001	M–W	0.474	<0.001	M–W	0.585	<0.001	M–W	0.654
Biochemical oxygen demand	mg/l	0.001	M–W	0.839	<0.001	M–W	0.179	0.002	M–W	0.027	<0.001	M–W	0.442
Chemical oxygen demand	mg/l	0.050	M–W	0.068	<0.001	M–W	0.395	0.051	T–test	0.039	<0.001	M–W	<0.001
Ammonia–nitrogen	mg/l	0.067	T–test	0.065	<0.001	M–W	0.001	0.002	M–W	0.461	<0.001	M–W	0.088
Nitrate–nitrogen	mg/l	0.007	M–W	0.092	<0.001	M–W	0.044	<0.001	M–W	0.168	<0.001	M–W	0.001
Ortho–phosphate–phosphorus	mg/l	0.009	M–W	0.056	0.001	M–W	0.002	0.001	M–W	0.179	<0.001	M–W	0.001
Aluminium	mg/l	0.011	M–W	0.001	<0.001	M–W	0.761	<0.001	M–W	0.008	0.001	M–W	0.002
Boron	mg/l	<0.001	M–W	<0.001	<0.001	M–W	0.045	<0.001	M–W	0.846	0.083	T–test	0.123
Calcium	mg/l	<0.001	M–W	0.001	<0.001	M–W	0.001	<0.001	M–W	0.008	0.001	M–W	0.606
Cadmium	mg/l	<0.001	M–W	<0.001	0.052	T–test	0.019	<0.001	M–W	0.003	<0.001	M–W	0.443
Chromium	mg/l	<0.001	M–W	<0.001	<0.001	M–W	0.012	<0.001	M–W	<0.001	<0.001	M–W	0.133
Copper	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	0.069
Iron	mg/l	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	0.058
Potassium	mg/l	0.517	T–test	0.220	0.225	T–test	0.687	<0.001	M–W	0.115	<0.001	M–W	0.674
Magnesium	mg/l	<0.001	M–W	0.001	<0.001	M–W	0.085	<0.001	M–W	0.576	<0.001	M–W	0.515
Manganese	mg/l	<0.001	M–W	<0.001	<0.001	M–W	0.011	<0.001	M–W	0.002	<0.001	M–W	0.280
Sodium	mg/l	0.061	T–test	0.696	0.555	T–test	0.587	0.005	M–W	0.550	0.005	M–W	0.118
Nickel	mg/l	<0.001	M–W	0.028	<0.001	M–W	0.001	<0.001	M–W	0.726	<0.001	M–W	0.184
Zinc	mg/l	<0.001	M–W	<0.001	<0.001	M–W	0.242	0.017	M–W	<0.001	<0.001	M–W	0.013

Note: HC, high concentration; LC, low concentration; SGW, synthetic greywater; T2, T6, T10, and T14, treatment systems with floating *P. australis* and ochre pellets; T4, T8, T12, and T16, treatment systems with ochre pellets only; Shapiro–Wilk (check for normality), normally distributed data if $p > 0.05$ using T–test and non–normally distributed data if $p < 0.05$ using Mann–Whitney U–test; p value, significantly different, if $p < 0.05$ and not significantly different if $p > 0.05$; M–W, Mann–Whitney U–test; NTU, nephelometric turbidity unit.

Table 7.4. (Continued).

(c) effects of contact time and pollutant loads on greywater remediation in treatment systems (T) contained floating *P. australis* and ochre pellets

		Effect of contact time						***Effect of pollutant loads***					
		HC–SGW (T2 & T10)			**LC–SGW (T6 & T14)**			**2–day contact time (T2 & T6)**			**7–day contact time (T10 & T14)**		
Parameter	***Unit***	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
pH	–	0.007	M–W	0.010	0.001	M–W	0.828	–	–	–	–	–	–
Redox potential	mV	0.008	M–W	0.018	<0.001	M–W	0.991	–	–	–	–	–	–
Turbidity	NTU	0.003	M–W	0.069	<0.001	M–W	0.058	<0.001	M–W	0.768	<0.001	M–W	0.648
Total suspended solids	mg/l	0.610	T–test	0.020	<0.001	M–W	0.010	<0.001	M–W	0.677	<0.001	M–W	0.601
Electrical conductivity	µS/cm	<0.001	M–W	0.893	<0.001	M–W	0.638	–	–	–	–	–	–
Dissolved oxygen	mg/l	0.474	T–test	0.007	0.706	T–test	0.570	0.165	T–test	0.829	0.170	T–test	0.282
Colour	Pa/Co	0.375	T–test	0.006	0.113	T–test	0.064	<0.001	M–W	0.883	<0.001	M–W	0.564
Temperature	°C	<0.001	M–W	0.738	<0.001	M–W	0.514	–	–	–	–	–	–
Biochemical oxygen demand	mg/l	0.125	T–test	0.493	<0.001	M–W	0.584	<0.001	M–W	0.264	<0.001	M–W	0.197
Chemical oxygen demand	mg/l	0.370	T–test	0.006	<0.001	M–W	0.760	<0.001	M–W	0.502	<0.001	M–W	0.123
Ammonia–nitrogen	mg/l	0.005	M–W	<0.001	<0.001	M–W	0.011	<0.001	M–W	0.066	<0.001	M–W	0.101
Nitrate–nitrogen	mg/l	0.002	M–W	0.031	<0.001	M–W	0.002	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Ortho–phosphate–phosphorus	mg/l	0.092	T–test	0.006	<0.001	M–W	0.055	0.001	M–W	0.361	<0.001	M–W	0.126
Aluminium	mg/l	<0.001	M–W	0.745	<0.001	M–W	0.001	<0.001	M–W	<0.001	<0.001	M–W	0.462
Boron	mg/l	<0.001	M–W	0.520	<0.001	M–W	0.001	<0.001	M–W	0.308	0.004	M–W	0.158
Calcium	mg/l	<0.001	M–W	0.081	0.004	M–W	<0.001	0.005	M–W	<0.001	<0.001	M–W	<0.001
Cadmium	mg/l	<0.001	M–W	0.493	0.001	M–W	0.910	0.046	M–W	0.001	0.017	M–W	0.008
Chromium	mg/l	<0.001	M–W	0.196	<0.001	M–W	0.404	<0.001	M–W	0.131	<0.001	M–W	0.298
Copper	mg/l	<0.001	M–W	0.402	<0.001	M–W	0.065	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Iron	mg/l	<0.001	M–W	0.485	<0.001	M–W	<0.001	<0.001	M–W	0.598	<0.001	M–W	<0.001
Potassium	mg/l	0.002	M–W	1.000	<0.001	M–W	1.000	<0.001	M–W	0.083	<0.001	M–W	0.141
Magnesium	mg/l	<0.001	M–W	0.903	<0.001	M–W	0.002	<0.001	M–W	<0.001	<0.001	M–W	0.373
Manganese	mg/l	<0.001	M–W	0.225	<0.001	M–W	0.371	<0.001	M–W	<0.001	0.001	M–W	0.909
Sodium	mg/l	0.204	T–test	0.820	0.078	T–test	0.714	0.031	M–W	0.004	0.005	M–W	0.002
Nickel	mg/l	<0.001	M–W	0.453	<0.001	M–W	0.026	<0.001	M–W	0.011	<0.001	M–W	0.002
Zinc	mg/l	<0.001	M–W	0.279	<0.001	M–W	0.227	<0.001	M–W	<0.001	<0.001	M–W	0.238

Note: HC, high concentration; LC, low concentration; SGW, synthetic greywater; Shapiro–Wilk (check for normality), normally distributed data if $p > 0.05$ using T–test and non–normally distributed data if $p < 0.05$ using Mann–Whitney U–test; p value, significantly different, if $p < 0.05$ and not significantly different if $p > 0.05$; M–W, Mann–Whitney U–test; NTU, nephelometric turbidity unit.

7.2.3. Dissolved Oxygen, Biochemical and Chemical Oxygen Demands

From the statistical analysis (Table 7.3), dissolved oxygen (DO), 5–day biochemical oxygen demand (BOD_5) and chemical oxygen demand (COD) decreased significantly with presence of *P. australis* in treatment systems of both types of SGWs (T1, T5, T9 and T13) compared with those values of inflow and compared to outflows of systems T3, T7, T11 and T15, respectively. In terms of DO, these findings confirmed the other published findings by previous researchers (Sooknah and Wilkie, 2004; Iamchaturapatr et al., 2007). However, it has been reported that the existence of macrophytes in wetlands has a slight effect on outflow DO concentrations, and significant effects of vegetation on BOD_5 removal have been reported in experimental wetlands (Tanner, et al., 1995). Furthermore, the BOD_5 removals (Figure 7.1) in vegetated wetlands were reported to be 15% higher than those removals in unvegetated wetlands (Burgoon et al., 1989), while COD removals (Figure 7.2) have been successfully reduced in vegetated horizontal flow wetlands compared to those removals in unvegetated wetlands (Akratos and Tsihrintzis, 2007).

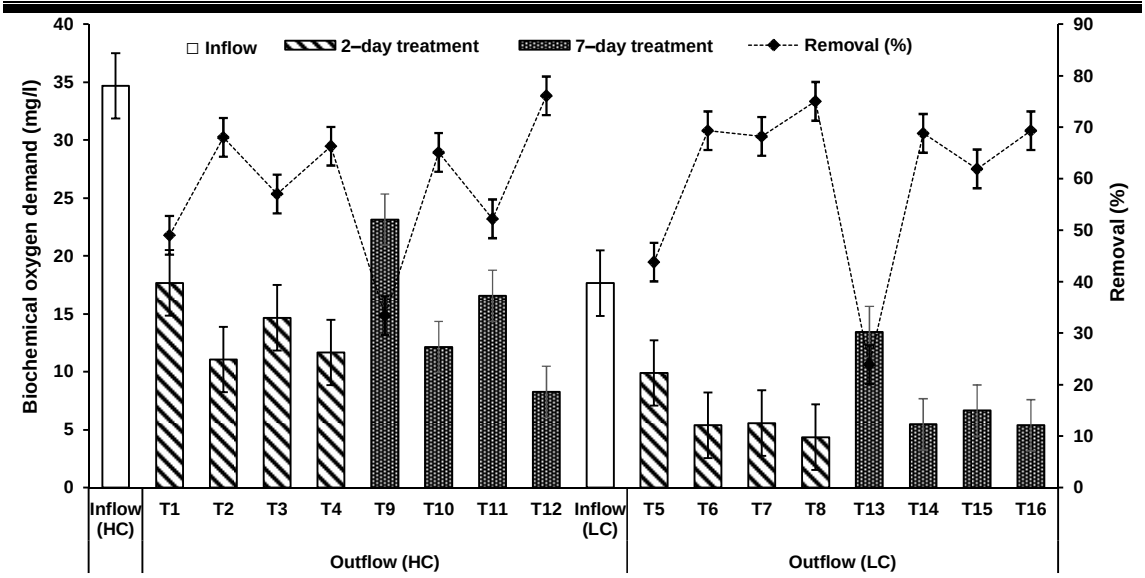


Figure 7.1. Overall performance of floating treatment wetlands in terms of biochemical oxygen demand removal.

The DO, BOD_5 and COD concentrations were recorded as significantly low in all treatment systems treating HC–SGW for both contact times, as well as systems using a combination of ochre pellets with *P. australis* (T2: 2–day and T10: 7–day), compared with those parameters of inflow (Table 7.3). Treatment of LC–SGW in systems using a combination of ochre pellets with *P. australis* showed significant decreases in DO and BOD_5 concentrations, but no significant changes in concentrations of COD in outflows of systems T6 (7–day) and T14 (7–day) when they were compared with those values of inflow. Similar effects on DO, BOD_5 and COD values were indicated when ochre pellets were present in combination with *P. australis* in systems T6 and T14 if they were compared to those values of outflows of systems T5 (2–day; only *P. australis*) and T10 (7–day; *P. australis*), respectively (Figures 7.1 and 7.2).

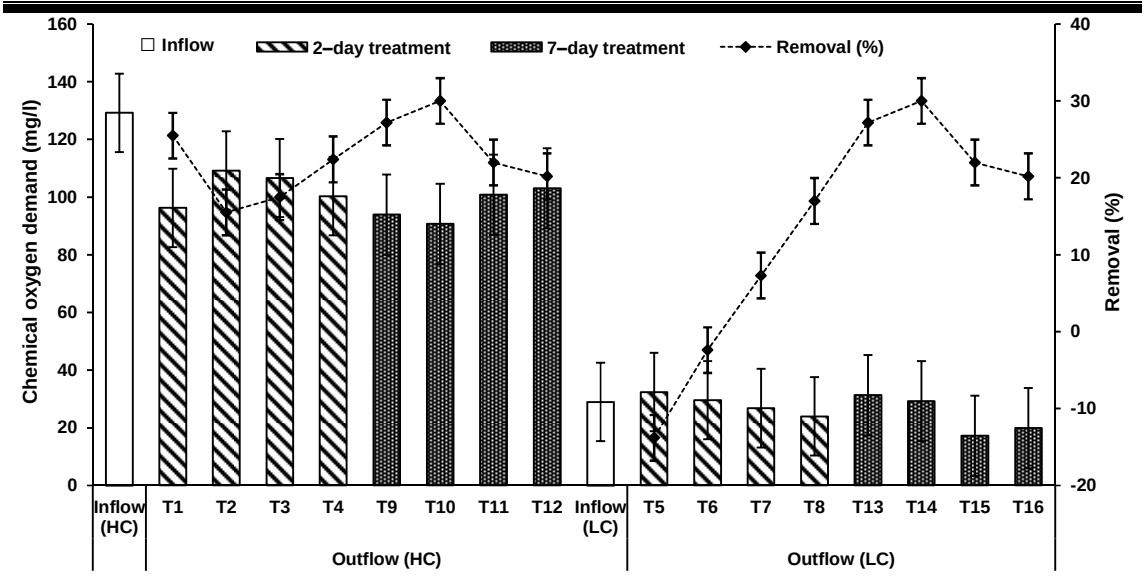


Figure 7.2. Overall performance of floating treatment wetlands in terms of chemical oxygen demand removal.

In addition, presence of cement–ochre pellets alone in treatment of SGW (T4, T8, T12 and T16) also resulted in significant decreases in DO, BOD_5 and COD compared with inflow and with outflows of systems T3, T7, T11 and T15, respectively (Figures 7.3 and 7.4). In particular, the significant reduction in DO and COD associated with the existence of ochre pellets is essentially due to the chemical oxidation processes involving elements such as Fe, Al and Ca in greywater (Dobbie et al., 2009; Littler et al., 2013).

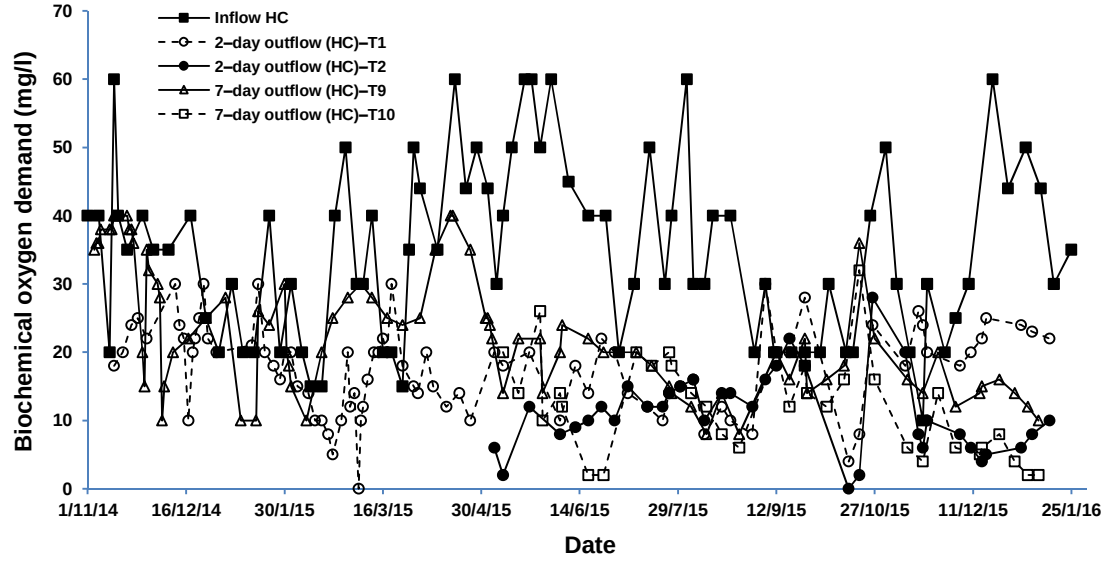


Figure 7.3. Effect of cement–ochre pellets and contact time on the variation of BOD_5 concentrations in high contamination (HC) synthetic greywater.

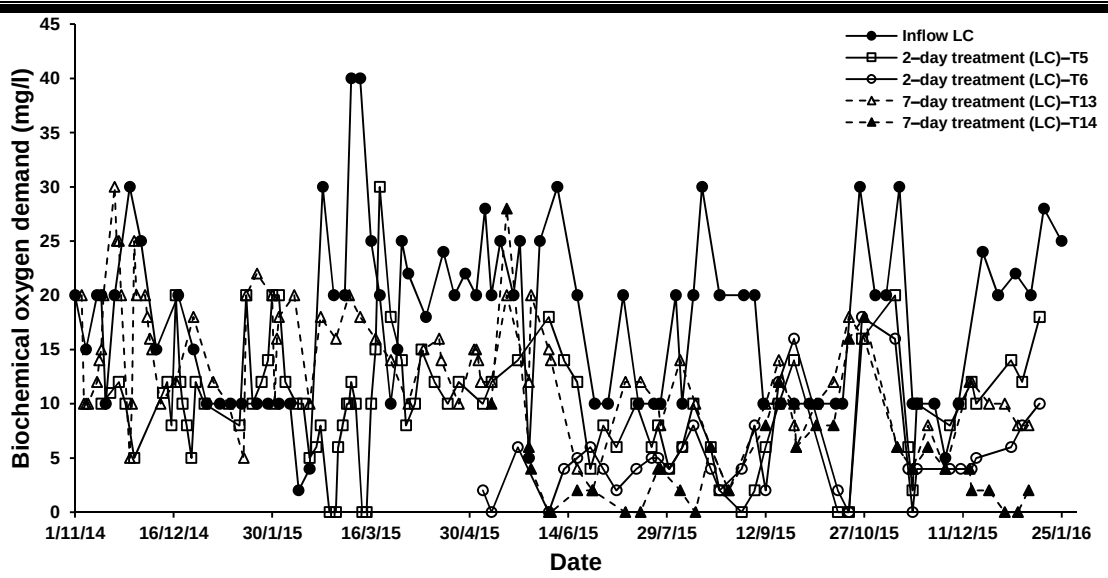


Figure 7.4. Effect of cement–ochre pellets and contact time on the variation of BOD_5 concentrations in low contamination (LC) synthetic greywater.

The DO, BOD_5 and COD values were not affected by increasing contact time in system T14 (7–day; combination of ochre pellets with *P. australis*) compared with those concentrations in the outflow of system T6 (2–day; combination of ochre pellets with *P. australis*), as shown in Table 7.4 and Figures 7.3–7.10. It was mentioned that increasing the contact time leads to an increase in DO, and this has also been confirmed in previous studies by Tanner et al. (1995). Furthermore, the increase of treatment time in vegetated floating wetlands reduced their performance in removing BOD_5 (Figures 7.3 and 7.4), which is indirectly required for the biological degradation of organic matter by microorganisms (Gross et al., 2007b; Vymazal and Kröpfelová, 2009), as shown in Figures 7.5 and 7.6.

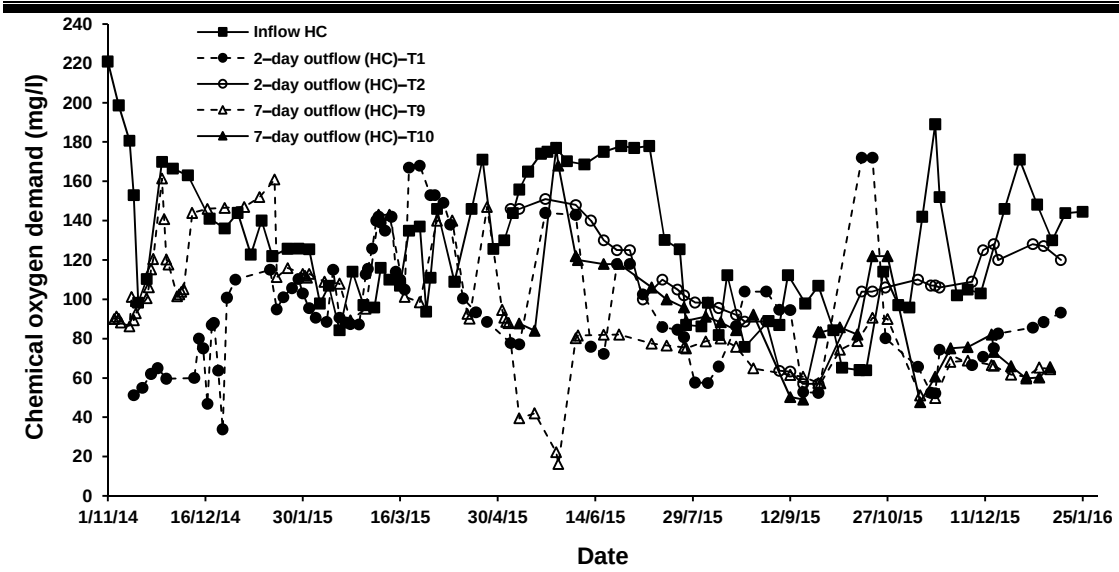


Figure 7.5. Effect of cement–ochre pellets and contact time on the variation of COD concentrations in high contamination (HC) synthetic greywater.

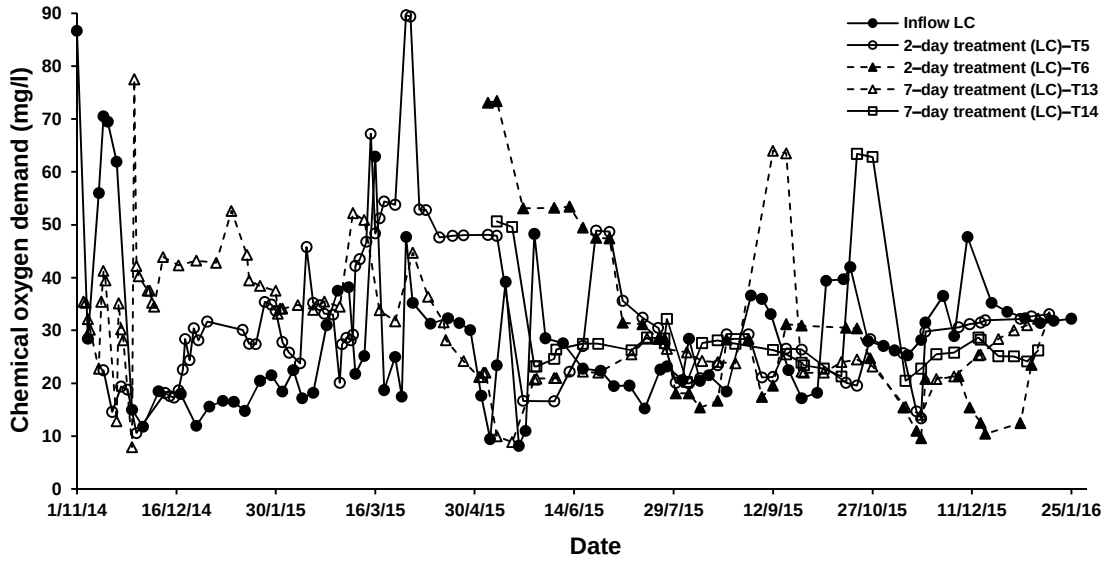


Figure 7.6. Effect of cement–ochre pellets and contact time on the variation of COD concentrations in low contamination (LC) synthetic greywater.

In addition, The DO, BOD_5 (Figures 7.7 and 7.8) and COD (Figures 7.9 and 7.10) values were not affected significantly by pollutant strengths of greywater when comparing systems treating HC–SGW using a combination of ochre pellets with *P. australis* (T2: 2–day and T10: 7–day) with those systems treating LC–SGW (T6: 2–day and T14: 7–day, respectively) at both contact times (Table 7.4).

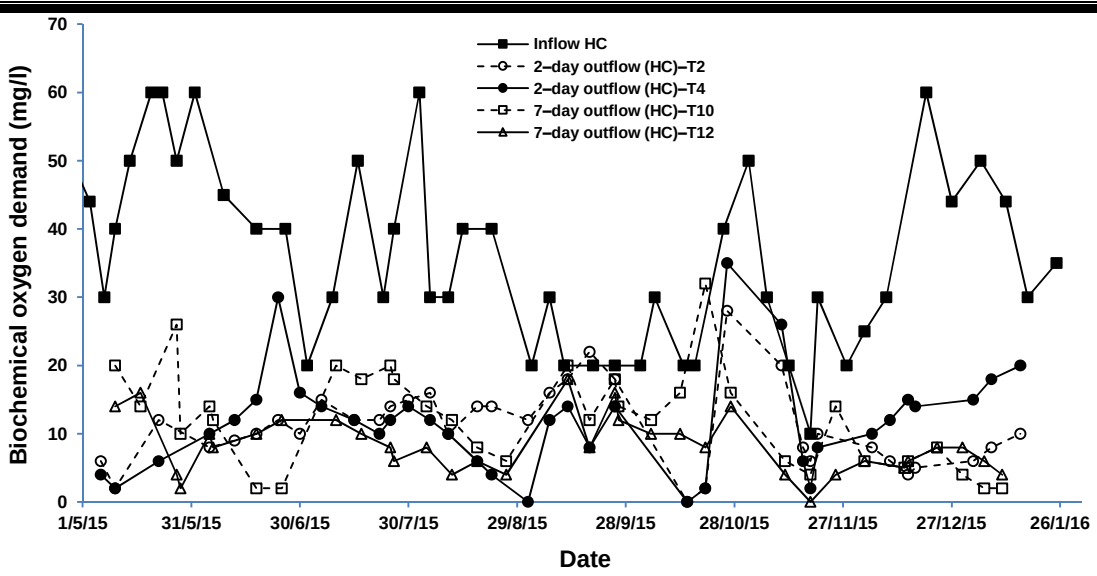


Figure 7.7. Effect of *P. australis* and contact time on the variation of BOD_5 concentrations in high contamination (HC) synthetic greywater.

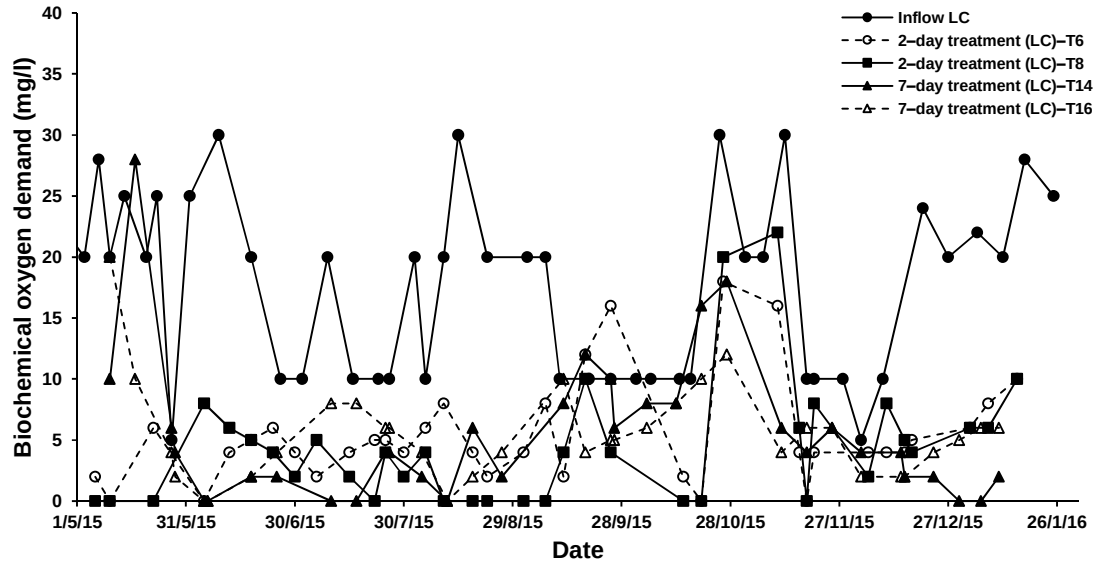


Figure 7.8. Effect of *P. australis* and contact time on the variation of BOD_5 concentrations in low contamination (LC) synthetic greywater.

Significant negative relationship correlations were observed between pH and BOD_5 ($r = -0.983$, $p < 0.001$). The strong negative correlation between pH and BOD_5 may occur through removal of organic matter in settling and biological processes (Hamersley and Howes, 2002), which consumes nutrients and carbonate components leading to an increase in pH due to the production of carbon dioxide for cell growth, and hydroxyl ions are left in excess (Wallace et al., 2016).

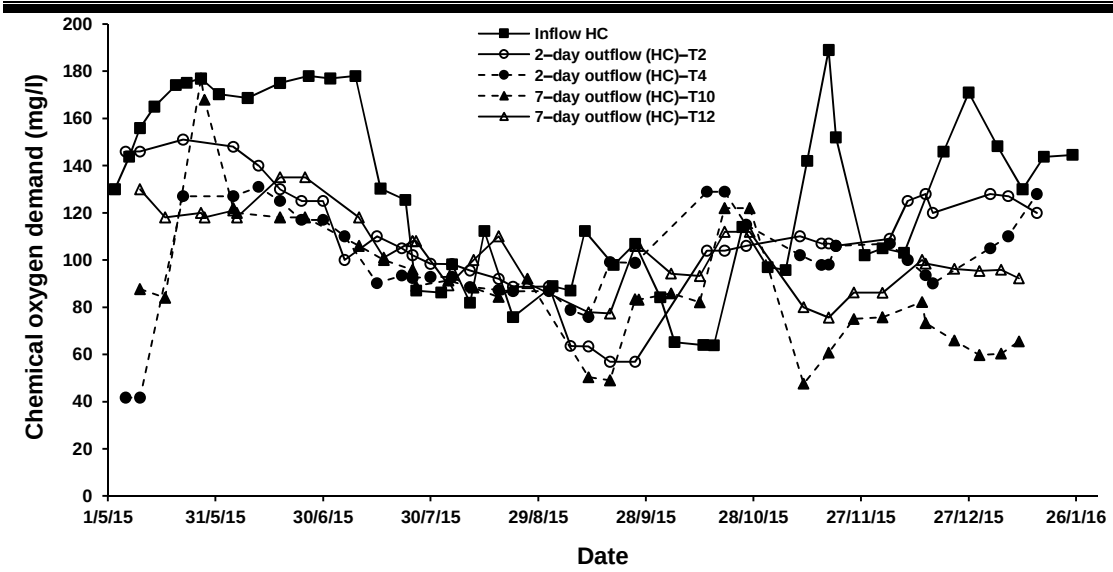


Figure 7.9. Effect of *P. australis* and contact time on the variation of COD concentrations in high contamination (HC) synthetic greywater.

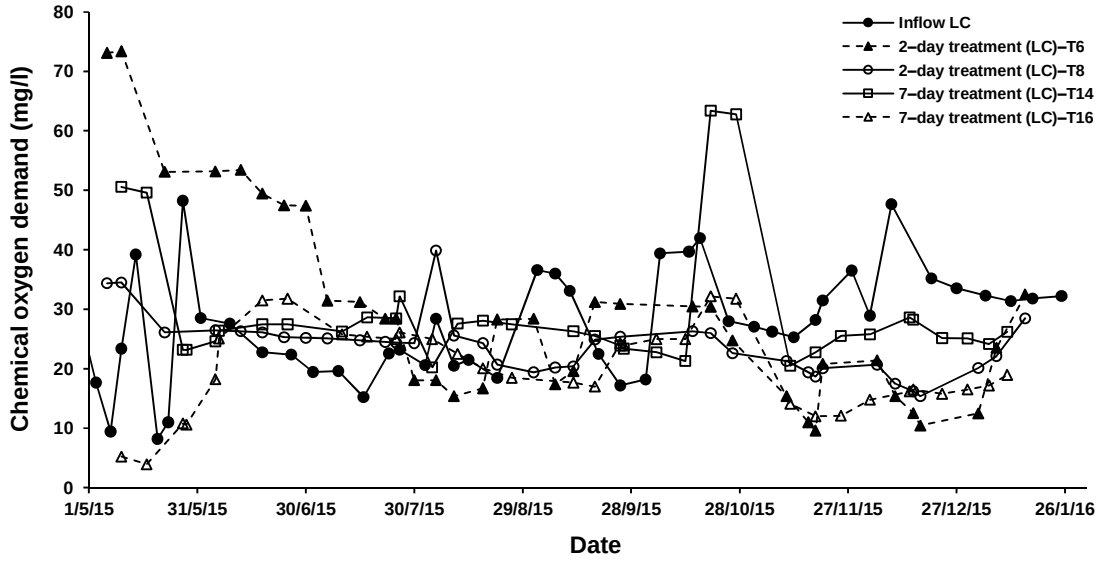


Figure 7.10. Effect of *P. australis* and contact time on the variation of COD concentrations in low contamination (LC) synthetic greywater.

However, there were considerable correlations observed between DO and COD which were negatively significant (r= -0.700, p = 0.036) in the treatment of LC–SGW and positively significant (r = 0.667, p = 0.050) for HC–SGW. Significantly ($p < 0.05$) negative correlations between DO and COD and/or BOD_5 were observed for wetlands with no limited oxygen supply, where aerobic degradation is governed by the amount of active organic matter during the chemical oxidation process and that which is available for the organisms as BOD_5 (Vymazal, 2005; 2010). For most types of greywater,

industrial wastewaters and storm water runoffs, DO correlates negatively with COD, because the supply of dissolved organic matter is not sufficient and aerobic degradation is not limited by DO (X. Li et al., 2009; Vymazal and Kröpfelová, 2009). In the case of significantly ($p < 0.05$) positive correlations of DO with COD, the sufficient supply of oxygen to aerobic *heterotrophic* bacteria will greatly increase the performance of aerobic biochemical oxidation (X. Li et al., 2009).

7.2.4. Nutrients (NH_4–N, NO_3–N and PO_4–P)

Although, there were no significant changes in concentrations of NH_4–N and NO_3–N, the considerable significant decreases of PO_4–P concentrations in treatment systems containing only SGW (T3, T7, T11 and T15) compared to values of inflow, were mainly due to either sedimentation and/or microorganism mechanisms (Figure 7.11). However, there was nutrient imbalance, in particular for LC–SGW, which was not sufficient for the organisms' survival and consequently limited the PO_4–P removal (Jefferson et al., 2001; Abed et al., 2017). For that reason, outflow treated SGW was agitated, in this study, before sampling to encourage solids to dissolve and/or re–suspend into the water column (Stewart et al., 2008; Van de Moortel et al., 2010; Tanner and Headley, 2011) in investigating the role of floating *P. australis* and cement–ochre pellets in treatment systems.

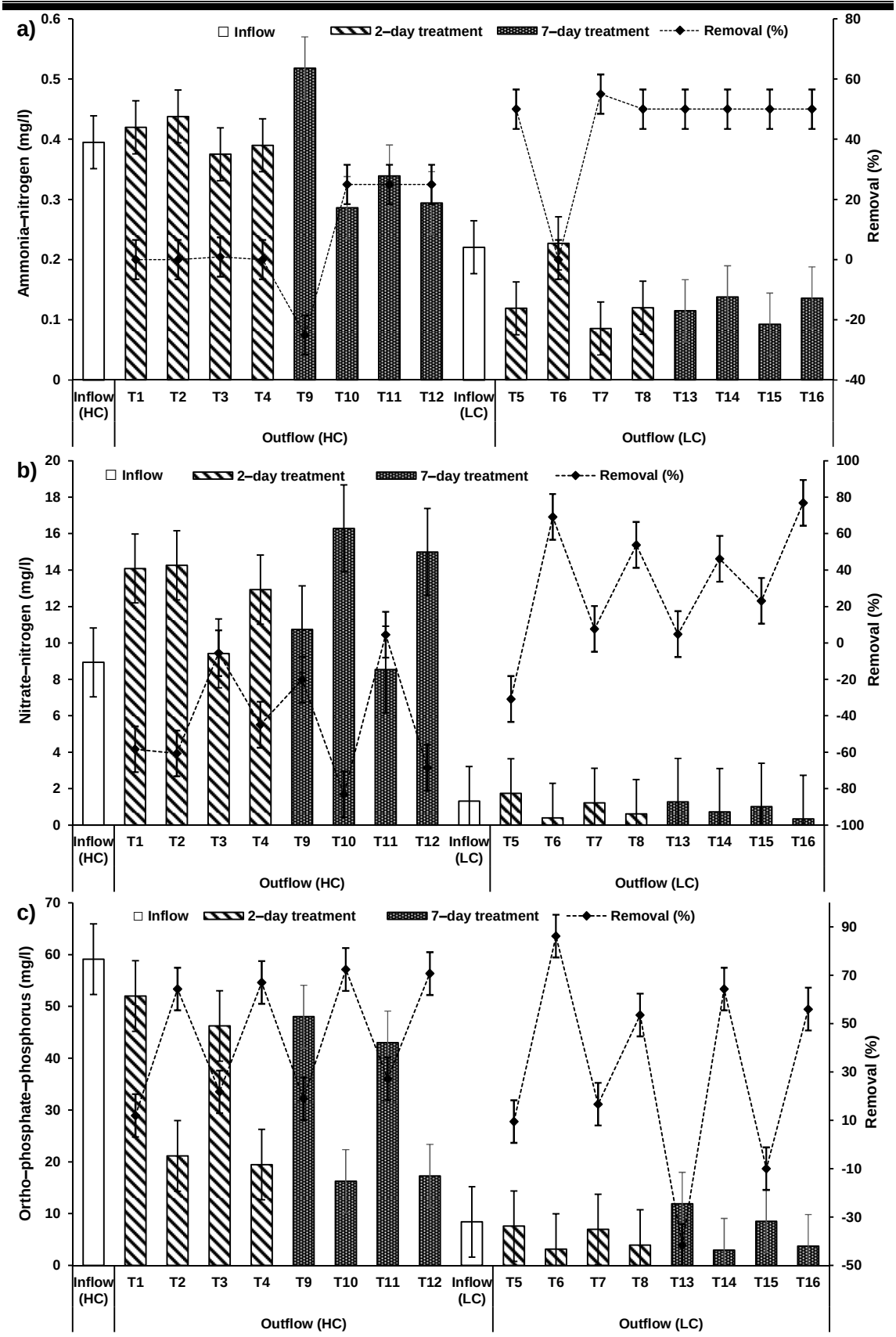


Figure 7.11. Overall performance of floating treatment wetlands with different operational parameters for treatment of both concentrations of synthetic greywater for 2– and 7–day contact times in order to remove; a) NH_4–N, b) NO_3–N, and c) PO_4–P.

The comparative statistical analysis (Table 7.3) of inflow with outflow of system T2 treating HC–SGW by combination of ochre pellets with floating *P. australis* for 2–day contact time (Figure 7.11) showed that concentration of NO_3–N was significantly higher, and PO_4–P was significantly lower than those concentrations of inflow, however, there was no significant change in NH_4–N concentration. Those effects were similar in comparing inflow with outflow HC–SGW from systems T1 (2–day; only *P. australis*) and T4 (2–day; only ochre pellets). Therefore, the significant decrease of PO_4–P concentration was linked to the effect of the presence of ochre pellets in combination with *P. australis* in system T2 compared to system T1 (Figure 7.12). However, there were no significant changes in NH_4–N and NO_3–N concentrations. In contrast, there was no significant effect on those parameters when *P. australis* was present in combination with ochre pellets in system T2 compared with system T4 (Table 7.4).

In systems using a combination of ochre pellets with floating *P. australis* treating LC–SGW (T6: 2–day and T14: 7–day contact time), concentrations of NO_3–N and PO_4–P were significantly lower than those of inflow, however, there were no significant changes in concentrations of NH_4–N in those systems compared to the NH_4–N value of the inflow (Figure 7.13). Those differences in mentioned parameters, with the exception of NH_4–N which significantly decreased here, were similar to the performance of systems T8 (2–day; only ochre pellets) and T16 (7–day; only ochre pellets) when compared to inflow (Table 7.3). Considering that in treatment systems treating SGW with only floating *P. australis*, volatilisation and plant uptake are primary removal mechanisms of NH_4–N removal in floating treatment wetland systems (Ferrara and Avci, 1982), the reduction of other nitrogenous forms such as NO_3–N and organic–N could cause the ammonia generation which may be behind the substantial increases of NH_4–N in some wetlands (Wallace et al., 2016).

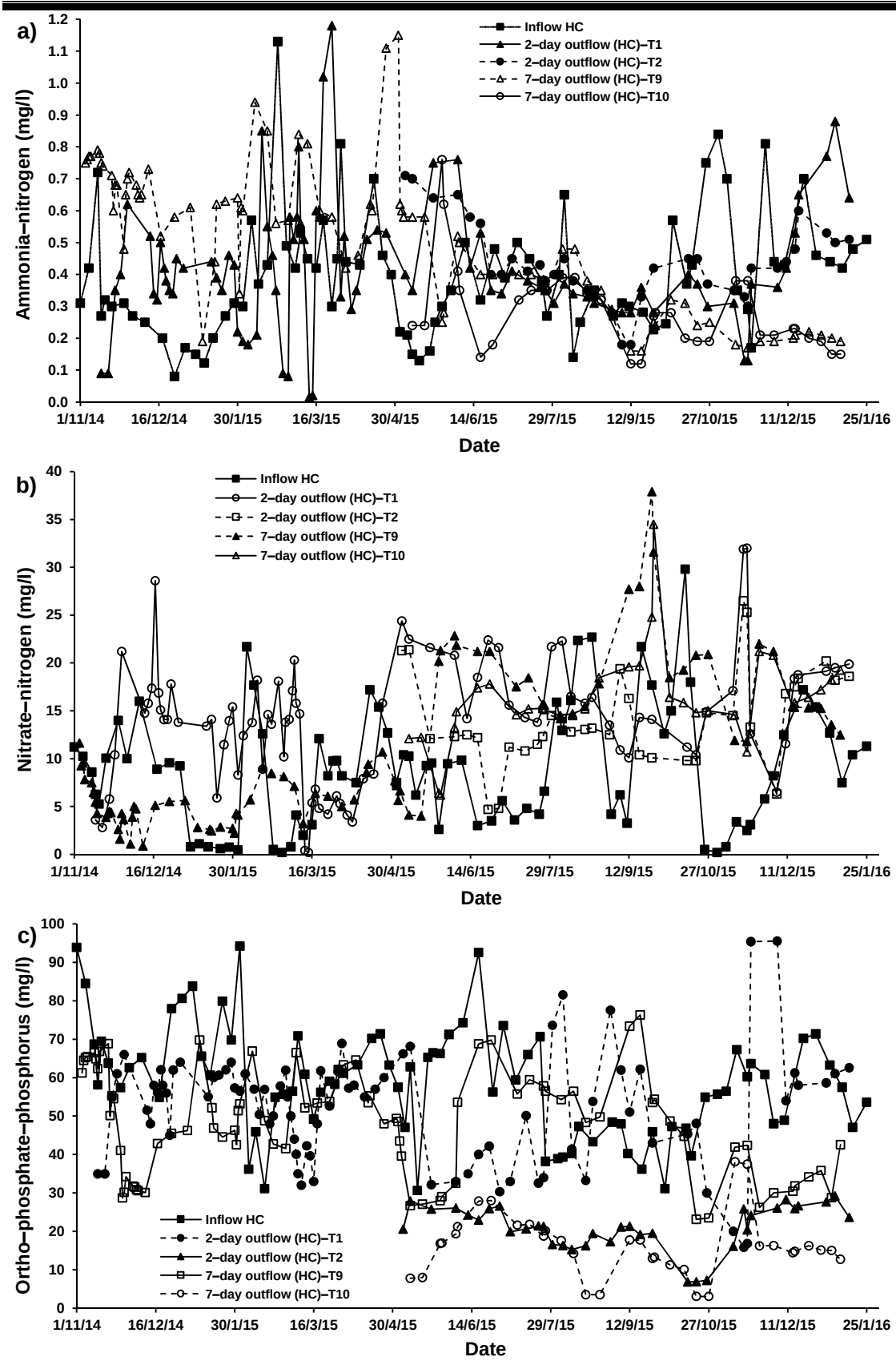


Figure 7.12. Effect of cement–ochre pellets and contact time on treatment of high contamination (HC) synthetic greywater in terms of the variation in concentrations of a) NH_4–N, b) NO_3–N, and c) PO_4–P.

Nitrogen components are present as NO_3 and/or NH_4 in the effluent. Nitrification and denitrification are natural metabolic processes of plants and associated microbes, including ammonium oxidation. The precipitation of PO_4–P is significantly reported as a removal mechanism in ponds, wetlands and similar systems (Ann et al., 1999; Peng et al., 2007), which might be a significant factor in PO_4–P removal. The uptake by algae and/or plants was also another likely factor in the PO_4–P reduction. However, this is a seasonal process that would likely have reversed during plant die–off (Wallace et al., 2016). Plants take–up phosphorus and nitrogen as an essential nutreints (Vymazal, 2010). However, the amount of phosphorus which is taken–up by plants is significantly lower than that for nitrogen (Brix, 1994).

A significant contribution in PO_4–P removal into FTWs is a result of the presence of ochre pellets in the treatment which is linked to either adsorption processes and/or precipitation processes, due to the high concentration of Ca, which converts the dissolved PO_4–P to insoluble substances under alkaline conditions (Heal et al., 2003; Dobbie et al., 2009), as shown in Figures 7.12 and 7.13.

Although the calcium phosphate precipitation was the dominant removal process for the cement–ochre pellets, it has been reported that it could be a minor proportion of removal associated with the ochre sludge (Littler et al., 2013). However, it was indicated that PO_4–P concentrations correlated significantly negatively with pH value ($r = -0.767$, $p = 0.016$) and Ca concentrations ($r = -0.783$, $p = 0.013$) in outflow of treatment systems. Furthermore, the outflow of the treatment systems with a combination of ochre pellets and *P. australis* showed that TSS correlated significantly negatively ($r = -0.717$. $p = 0.030$) with PO_4–P and significantly positively ($r = 0.717$, $p = 0.030$) with NO_3–N.

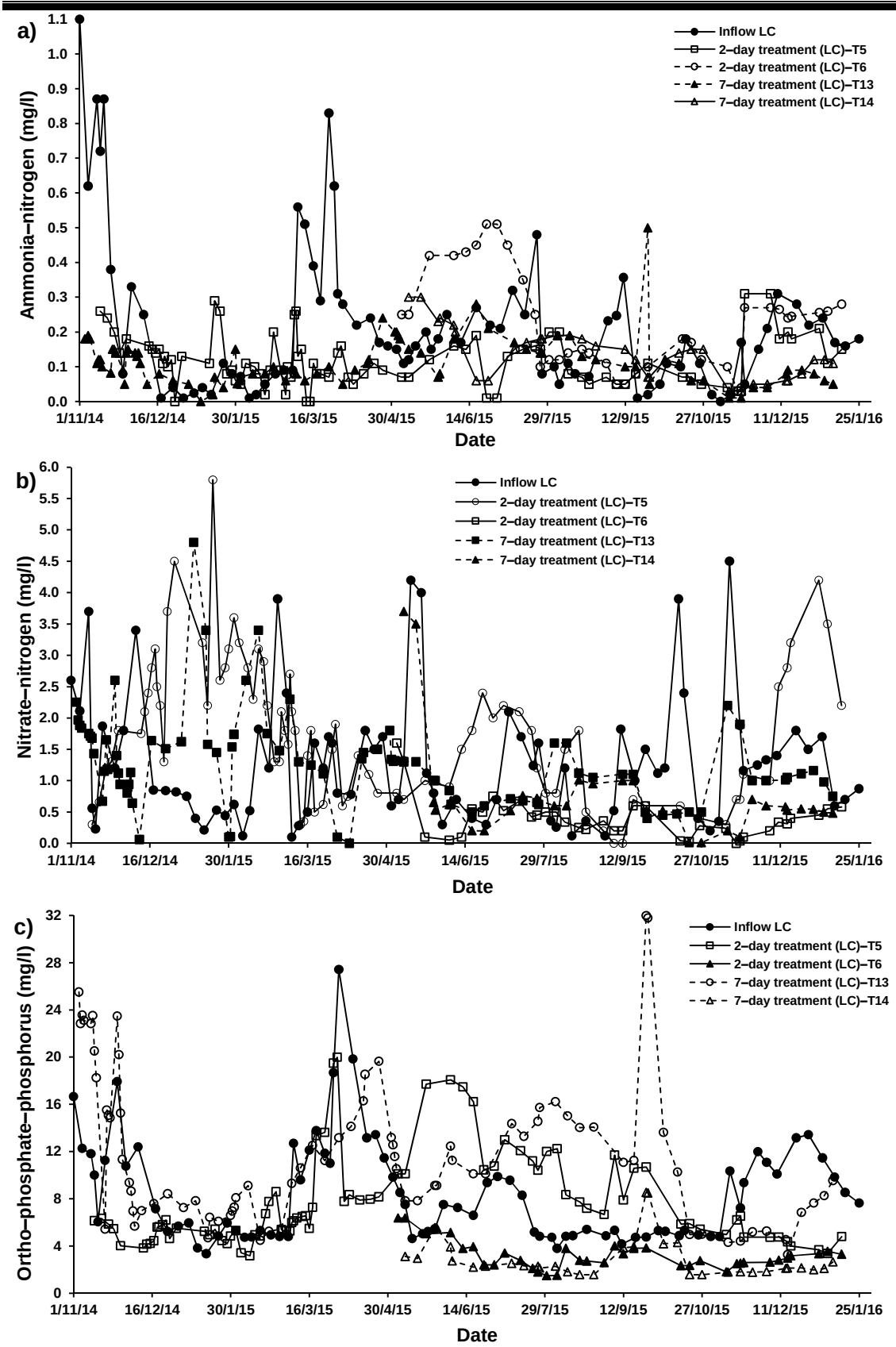


Figure 7.13. Effect of cement–ochre pellets and contact time on treatment of low contamination (LC) synthetic greywater in terms of the variation in concentrations of a) NH_4–N, b) NO_3–N, and c) PO_4–P.

After 7–days of treating HC–SGW in systems T10 (ochre pellets with *P. australis*) and T12 (only ochre pellets), the concentrations of NH_4–N and PO_4–P were decreased significantly, with significant increase in NO_3–N concentration noted, compared to those parameters of inflow (Table 7.3). Furthermore, the outflow of system T1 (only *P. australis*) showed the same pattern in terms of NH_4–N and PO_4–P, with no significant effect on NO_3–N concentration, when compared with inflow. Also in 7–day contact time of treatment of HC–SGW, the presence of ochre pellets in combination with *P. australis* in system T10 significantly increased NH_4–N and PO_4–P concentrations compared with the outflow of system T9 (only *P. australis*). However, there was no significant effect on those values from the presence of *P. australis* in combination with ochre pellets in system T10 compared with system T12 (only ochre pellets).

Regarding the increases in NO_3–N concentrations, it was claimed that greywater is nutrient–deficient, which impacts on the treatment efficiency (Jefferson et al., 2001; 2004). Therefore, nitrification is not generally considered an important mechanism in FTWs due to the lack of nitrifying bacteria in the facultative zone and ready uptake by plants (Ferrara and Avci, 1982), and was not expected to contribute significantly in ammonia conversion (Wallace et al., 2016). Therefore, those technologies are not efficient for removing nitrogenous compounds, particularly NO_3−N, because most greywaters lack biodegradable organic matter, which can be efficiently utilised by denitrifying bacteria (Xiong et al., 2012; Cao et al., 2016).

Therefore, presence of ochre pellets in combination with *P. australis* for treatment of LC–SGW significantly decreased the concentrations of NO_3–N and PO_4–P of outflow from systems T6 (2–day) and T14 (7–day), with no significant effect on NH_4–N concentrations, when compared with those values of outflow from systems with only *P. australis*, T5 (2–day) and T13 (7–day), respectively (Table 7.4 and Figure 7.13).

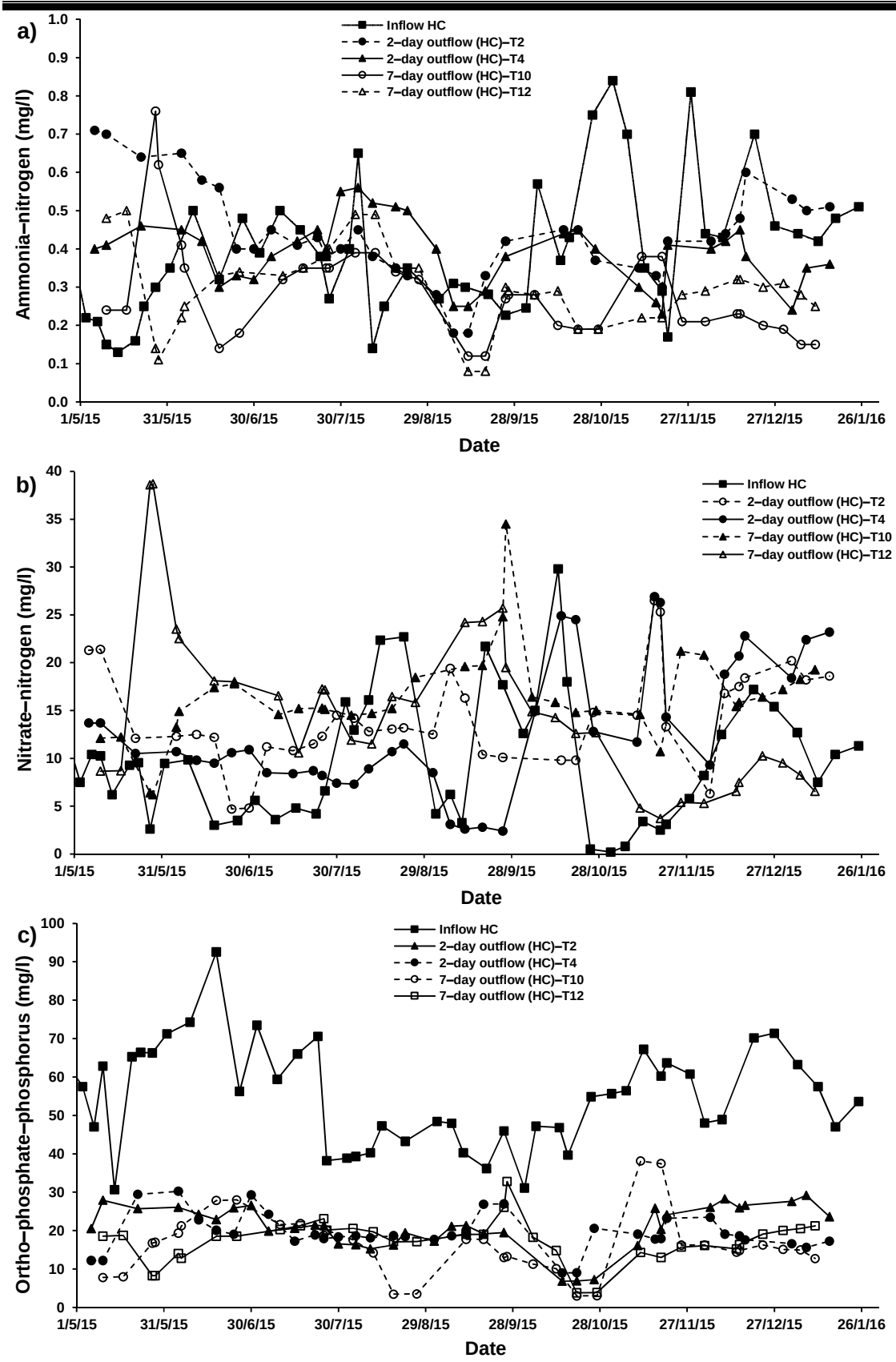


Figure 7.14. Effect of *P. australis* and contact time on treatment of high contamination (HC) synthetic greywater in terms of the variation in concentrations of a) NH_4–N, b) NO_3–N, and c) PO_4–P.

In addition, increasing contact time of treatment of HC–SGW (Figure 7.14) in systems using a combination of ochre pellets with *P. australis* significantly decreased the concentrations of NH_4–N and PO_4–P, with significant increase in NO_3–N concentration compared to the system of 2–day contact time of treatment. Moreover, increasing contact time of treatment of LC–SGW (Figure 7.15) in system T14 significantly decreased NH_4–N, and increased NO_3–N concentrations, with no significant effect on PO_4–P concentrations, when compared with outflow of system T6 (Figures 7.12–7.15). Removals of NO_3–N in systems treating LC–SGW with a combination of ochre pellets and *P. australis* were significantly higher than those removals in treatment of HC–SGW, however, no significant differences in removals of NH_4–N and PO_4–P were observed (Table 7.4).

However, it has been reported that cement–ochre pellets are able to continue removing phosphorus in experiments of up to seven days, whereas the unpelletised ochre sludge approached an equilibrium in a phosphate solution after about 60 minutes of contact time (Littler et al., 2013). Therefore, oxides and/or hydroxides of Fe and Al released from ochre pellets, and compounds of Ca had the ability to adsorb phosphorus (Heal et al., 2005). Phosphorus concentrations can decrease within a few minutes in non–agitated conditions (McHaffie et al., 2001). Therefore, an increase in contact time does not necessarily lead to a decrease in PO_4–P, if ochre pellets are present (Table 7.4).

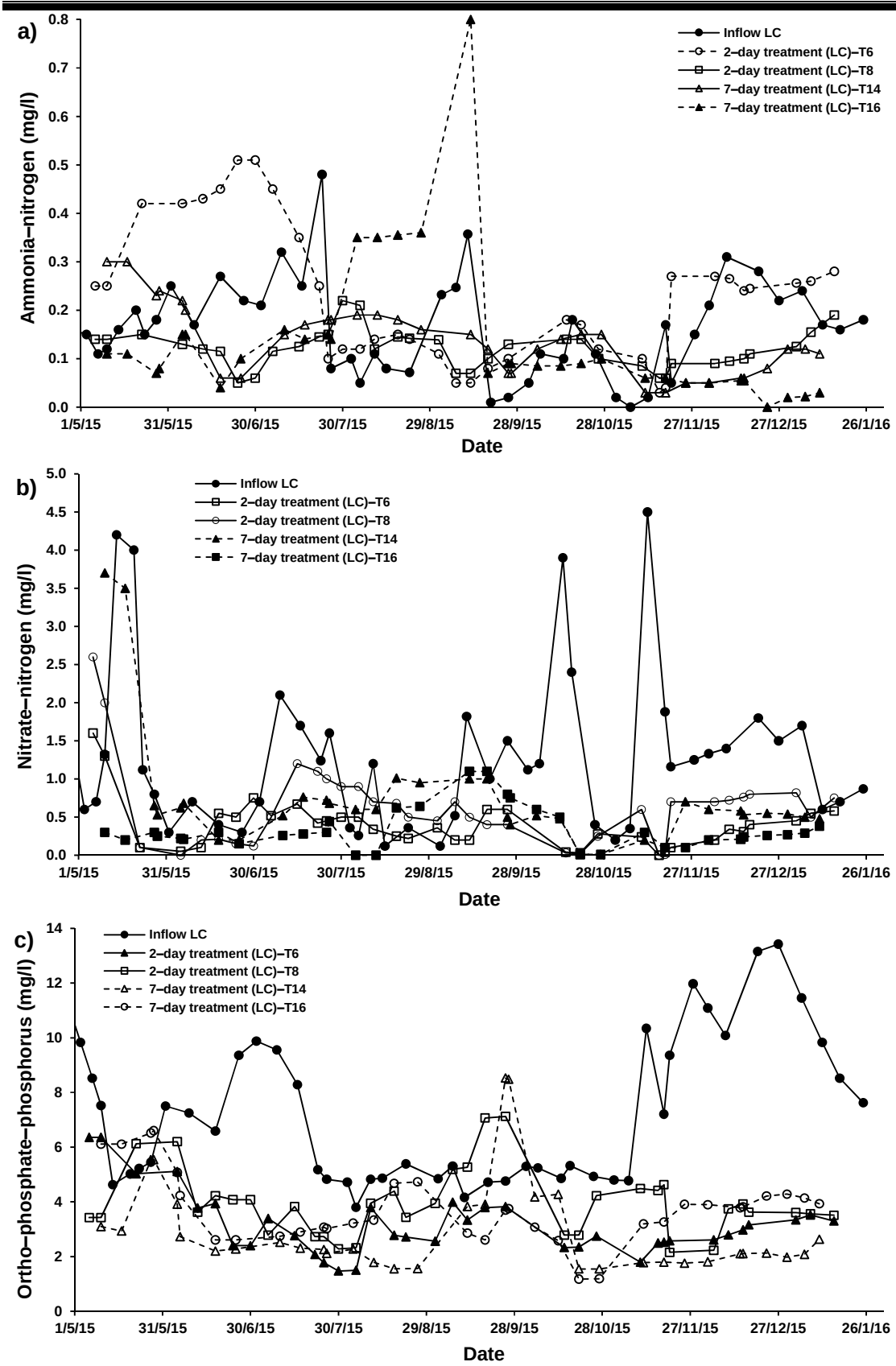


Figure 7.15. Effect of *P. australis* and contact time on treatment of low contamination (LC) synthetic greywater in terms of the variation in concentrations of a) NH_4–N, b) NO_3–N, and c) PO_4–P.

7.2.5. Trace Elements in Greywater

Trace element concentrations of inflow and outflow SGWs are shown in Tables 7.1 and 7.2, respectively. In the system treating HC–SGW by a combination of ochre pellets and floating *P. australis* for 2–day contact time (T2), the outflow exhibited significant decreases in concentrations of B, Cd, Cr, Cu, Fe, Mg, Mn, Ni and Zn compared with those contents of inflow (Figure 7.16). However, a significant increase in concentrations of Ca, and no significant changes in Al, K and Na, were observed, compared to inflow HC–SGW. Similar significant changes were indicated in outflow of system T1 (HC–SGW; 2–day; only *P. australis*), with the exception that Al and K showed significant decreases and Mg did not change significantly compared to those concentrations of inflow. System T4 (HC–SGW; 2–day; only ochre pellets) showed significant increases in contents of Ca and Cu, significant decreases in B, Mg, Mn and Ni, and no significant differences in other elements compared to those values of inflow. In contrast, comparison between inflow and outflow of system T3 (2–day; only HC–SGW) showed no significant differences in terms of the concentrations of Al, B, Cd, Cu, Fe, Mg, Na, Ni and Zn (Table 7.3 and Figure 7.16).

In FTWs, the trace elements might be removed by corporation processes such as settling, sedimentation, sorption, precipitation, cation exchange, photo–degradation, phytoaccumulation, biodegradation, microbial activity and plant uptake. However, it is difficult to determine the actual reactions which take place in FTWs (Collins et al., 2005; Borne et al., 2014)

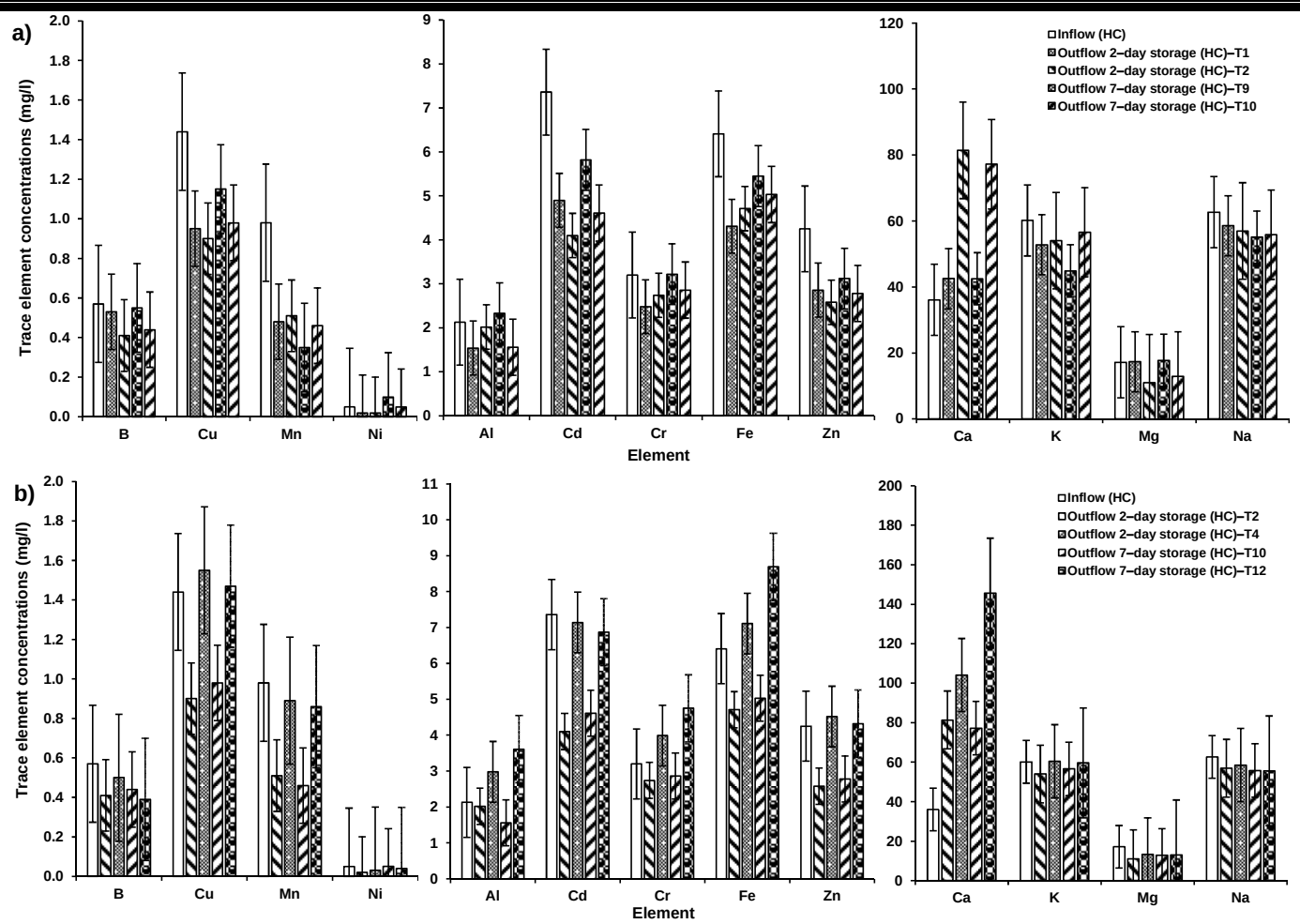


Figure 7.16. Variation in trace element concentration in treatment of high pollutant concentration (HC) synthetic greywater by floating treatment wetlands under the effect of a) cement ochre pellets and contact time, and b) *P. australis* and contact time.

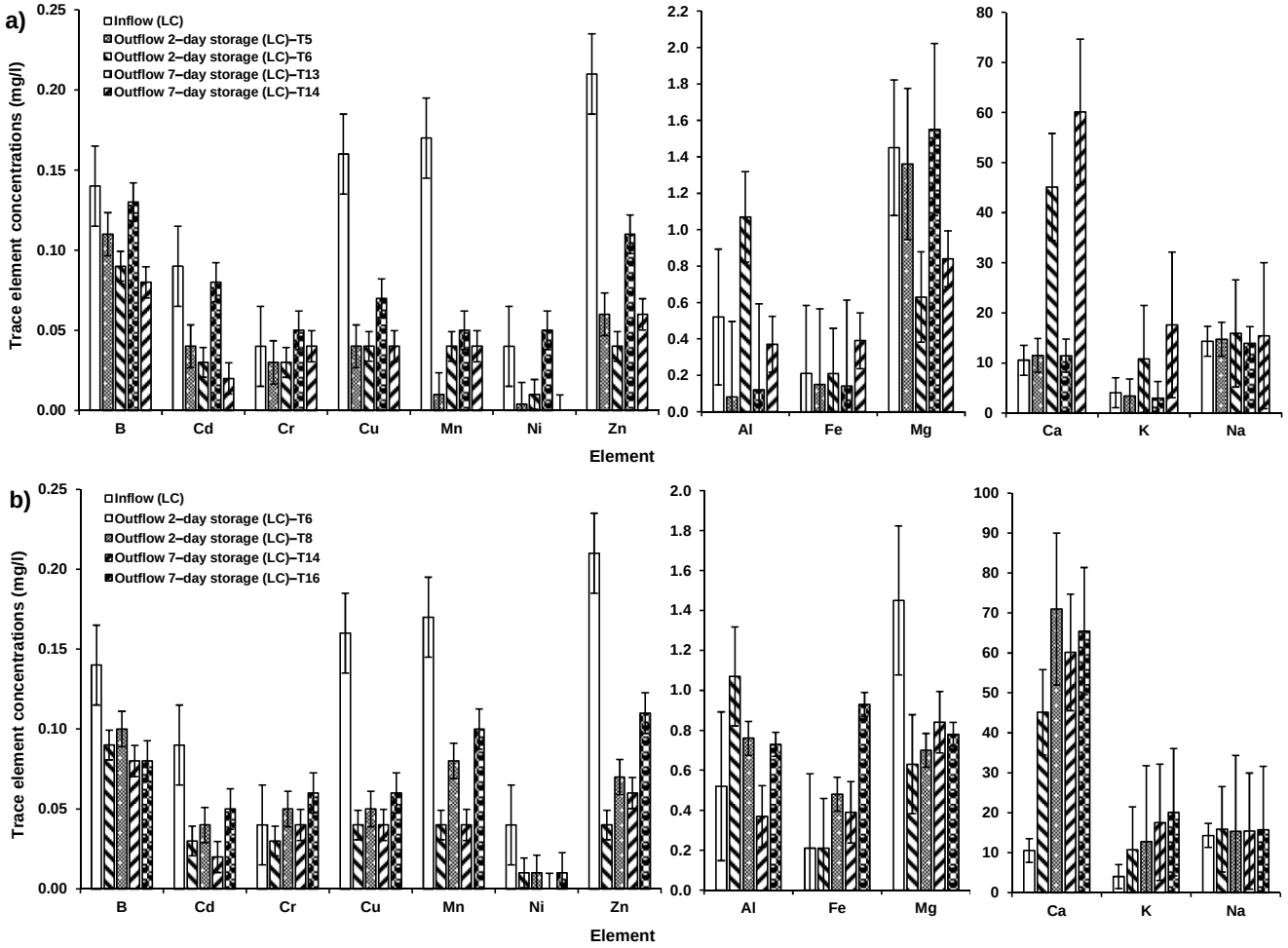


Figure 7.17. Variation in trace element concentration in treatment of low pollutant concentration (LC) synthetic greywater by floating treatment wetlands under the effect of a) cement ochre pellets and contact time, and b) *P. australis* and contact time.

The comparative analysis between outflows of systems T1 and T2 showed that the presence of ochre pellets in combination with *P. australis* in system T2 significantly decreased B and Mg while increasing Al, Ca and Fe contents in outflow. Those significant increases were linked to the high contents of Al, Ca and Fe in cement–ochre pellets which were released into the water column (Heal et al., 2005), as shown in Table 7.4. On the other hand, comparison between outflows of systems T2 and T4 revealed the effects of *P. australis*, when used in combination with ochre pellets in system T2, were significant decreases in the concentrations of all considered elements (Table 7.3).

In treatment of HC–SGW for 7–day contact time by a combination of ochre pellets and floating *P. australis*, in the outflow of system T10 concentrations of Al, B, Cd, Cu, Fe, Mg, Mn and Zn were still decreased significantly with significant increase in Ca content compared to those concentrations of inflow (Figure 7.16). However, there were no significant changes in contents of Cr, K, Na and Ni in that comparison, which reveals that a balance condition, in terms of those elements, occurred in that system (Littler et al., 2013).

For 7–day contact time, the ochre pellets with floating *P. australis* in system T10 (HC–SGW) significantly affected the removal of Al, B, Cd, Cr, Cu, Mg, Ni and Zn compared with those concentrations in the outflow from system T9. While, floating *P. australis* in combination with ochre pellets in system T10 contributed significantly to the decrease the concentrations of Al, Ca, Cd, Cr, Cu, Fe, K, Mg, Mn and Zn compared with the outflow of system T12. However, system T9 (7–day; HC–SGW; only *P. australis*) showed significant removals of Cu, K, Mn, Ni and Zn compared to inflow (Figure 7.17). System T12 (7–day; HC–SGW; only ochre pellets) removed B and Mg significantly compared to their inflow levels. In Table 7.4, the role of *P. australis* was very clear in terms of its adsorbing the dissolved metals; because the roots and rhizomes are not

planted in soils, they are forced to acquire their nutrition directly from the water column, which may enhance rates of nutrient and element uptake into biomass (Tanner and Headley, 2011). In parallel, dissolved elements in greywater, such as Al, Ca and Fe, might be adsorbed chemically to replace vacant places which were occupied by the over saturated elements in ochre pellets and then remobilised to the water column (Sibrell et al., 2009; Sapsford et al., 2015).

In treatment of LC–SGW by a combination of ochre pellets and floating *P. australis* for 2–day contact time in system T6, the concentrations of B, Cd, Cu, Mg, Mn, Ni and Zn were significantly decreased compared with those of inflow. However, there were significant increases in contents of Al, Ca, Fe and K indicated in the outflow of system T6 compared with inflow. Similar significant changes, as in system T6, were observed in outflows of systems T5 (2–day; LC–SGW; only floating *P. australis*) and T8 (2–day; LC–SGW; only ochre pellets), with the exception of significant decreases in Al, Fe and K for system T6, compared to those values of inflow (Figure 7.17). So, presence of ochre pellets in combination with floating *P. australis* in system T6 had a significant effect on the decrease in the concentrations of B, Cd and Zn, and the increase in the concentrations of Al, Ca, K, Mn and Ni compared to the outflow of system T5. Furthermore, the presence of floating *P. australis* contributed significantly to remove B, Ca, Cd, Cr, Cu, Fe, Mn, Ni, and Zn compared with the outflow from system T8 (Table 7.4 and Figure 7.17).

Although the water is stagnant in FTWs, settled particle solids, associated with trace elements and other pollutants, may be transported from the water to the ochre solids or biomass or vice versa (Kadlec and Knight, 1996; Stewart et al., 2008). However, using pelletised ochre solids in those FTWs promises an adsorption and sequestration of trace

elements, heavy metals in particular, through compositing those in safe form to become part of the geological cycle (Dobbie et al., 2009, Littler et al, 2013).

In 7–day contact time of treatment of LC–SGW with a combination of ochre pellets and floating *P. australis* in system T14, significant decreases in contents of B, Cd, Cu, Mg, Mn, Ni and Zn with associated significant increases in contents of Ca, Fe and K were indicated, compared with those contents of inflow. The same significant performance as in the outflow of T14, was noted in the outflow of system T13 (7–day; LC–SGW; only floating *P. australis*), but contents of Al, Fe, K, were lower, and content of Mg and Ni were higher than those of inflow. Furthermore, significant changes in the outflow of system T16 (7–day; LC–SGW; only ochre pellets) were noted, similar to those changes in the outflow of system T14, except there was a significant increase in Al content in the outflow of the former system (T16) compared with inflow (Table 7.3 and Figure 7.17). Therefore, presence of ochre pellets in combination with *P. australis* in system T14 had a significant effect on the decrease in B, Mg and Zn, and on the increase in Al, Ca, Fe and K compared with those contents of the outflow from system T13. Although there were insignificant decreases in concentrations of most elements, the presence of *P. australis* in that system (T14) significantly affected the decrease in Al and Zn compared to the outflow of system T16 (Tables 7.3 and 7.4).

However, there were no significant effects of increasing contact time of treatment of HC–SGW by a combination of ochre pellets with floating *P. australis* in system T10 compared with outflow of system T2 (2–day; HC–SGW; ochre pellets with floating *P. australis*). In treatment of LC–SGW with a combination of ochre pellets and floating *P. australis* in system T14, increasing contact time significantly decreased Al, B and Ni, while it increased Ca, Fe and Mg significantly compared with outflow of system T6 (2–day; LC–SGW; ochre pellets with floating *P. australis*). Some element concentrations

increased in the outflows of the treatment systems without the presence of ochre pellets, since all systems were subjected to the impacts of natural forces such as precipitation. It is also likely that contaminants such as dust particles and plant leaves entered the ponds (Mufarrege et al., 2010; Tanner and Headley, 2011; Borne et al., 2014).

In comparisons between removals of trace elements by a combination of ochre pellets and floating *P. australis* from HC–SGW and LC–SGW (T2 and T6; at 2–day: and, T10 and T14; at 7–day), the removals of Al, Ca and Na were higher in system T2 (HC–SGW) and removals of Ca, Fe and Na were higher in system T10 (HC–SGW) compared to systems T6 and T14, respectively. While, removals of Cd, Cu, Mg, Mn, Ni and Zn were higher in system T6 (LC–SGW) and removals of Cd, Cu and Ni were higher in system T14 (LC–SGW), compared to systems T2 and T10, respectively (Table 7.4).

7.3. Trace Elements Accumulated in Ochre pellets

The concentrations of trace elements which were in contents of raw ochre sludge and pelletised ochre before use in the treatment of SGW are illustrated in Table 7.5. At the end of the treatment, an elementary analysis of the ochre pellets for the accumulation of trace elements was carried out and results are shown in Table 7.6.

Table 7.5. Trace element concentrations (mg/kg) detected in digested samples of raw ochre sludge and pellets produced from adding Portland cement to ochre sludge.

Element	*Raw sludge ochre*					*Cement–ochre pellets*				
	n	Mean	SD	Min	Max	n	Mean	SD	Min	Max
Aluminium	18	177.8	124.74	<0.001	345.7	18	13696.4	686.05	12448.4	14941.7
Boron	15	37.0	37.71	5.9	109.3	15	36.9	16.22	17.5	62.8
Calcium	21	25677.4	2733.47	19218.7	30147.8	21	268527.3	11891.67	245019.6	290052.8
Cadmium	16	8.7	11.41	<0.001	33.9	18	4.5	6.31	<0.001	18.9
Chromium	27	22.5	32.0	<0.001	112.0	27	47.5	24.99	17.9	109.1
Copper	27	95.7	127.98	<0.001	439.1	27	74.8	59.27	26.3	216.2
Iron	24	470458.5	79408.35	336308.0	578228.0	24	224603.5	10102.35	204262.0	240776.8
Magnesium	27	286.6	96.66	123.7	543.2	27	4859.0	338.98	3932.9	5281.4
Manganese	27	4276.3	720.18	3184.7	5430.5	27	2171.8	131.56	1951.6	2492.9
Nickel	16	15.8	10.48	2.3	34.4	17	15.5	10.44	0.7	32.8
Zinc	24	70.4	86.29	<0.001	394.0	24	447.7	149.16	216.3	783.1

Note: n, number of tested samples; SD, standard deviation; Min, minimum; Max, maximum.

Table 7.6. Trace elements (mg/kg) accumulated in cement–ochre pellets after the treatment period of high concentration synthetic greywater (HC–SGW) and low concentration synthetic greywater (LC–SGW) in mesocosm–scale treatment systems (T) at (a) 2–day contact time and (b) 7–day contact time.

(a) 2–day contact time

HC–SGW

	Cement–ochre pellets in T2						Cement–ochre pellets in T4					
Element	n	Mean	SD	Min	Max	Accumulation (%)	n	Mean	SD	Min	Max	Accumulation (%)
Aluminium (Al)	18	15453.0	1226.75	13359.0	17573.7	12.8	18	14589.7	1153.27	12352.2	16197.7	6.5
Boron (B)	15	95.3	16.42	73.3	127.9	158.1	15	82.0	17.80	55.5	115.9	122.1
Calcium (Ca)	21	225571.9	12870.26	201974.0	248470.8	-16.0	21	214795.4	12783.36	190833.2	231786.0	-20.0
Cadmium (Cd)	12	183.3	59.18	123.9	279.3	4005.7	14	237.0	89.84	26.4	334.2	5208.6
Chromium (Cr)	27	286.0	228.90	52.0	665.8	501.5	27	121.5	74.45	41.3	277.4	155.6
Copper (Cu)	27	156.1	80.44	69.9	331.6	108.7	27	118.7	69.27	52.8	279.8	58.7
Iron (Fe)	24	250273.6	16905.83	217753.6	277509.2	11.43	24	235900.8	18729.34	198789.6	263774.4	5.0
Magnesium (Mg)	27	8039.4	757.51	6064.0	9071.8	65.5	27	7999.2	755.80	5994.7	8759.9	64.6
Manganese (Mn)	27	2493.9	195.09	2152.1	2963.2	14.8	27	2443.6	230.23	2024.0	2894.8	12.5
Nickel (Ni)	12	18.7	11.36	5.3	43.6	20.5	21	28.2	18.74	2.4	58.1	81.8
Zinc (Zn)	24	593.4	157.11	367.7	1019.2	32.5	24	546.0	160.59	334.6	917.9	22.0

LC–SGW

	Cement–ochre pellets in T6						Cement–ochre pellets in T8					
Element	n	Mean	SD	Min	Max	Accumulation (%)	n	Mean	SD	Min	Max	Accumulation (%)
Aluminium (Al)	18	16651.9	1307.90	14055.0	18726.7	21.6	18	16867.7	1114.87	14944.1	18942.3	23.2
Boron (B)	15	43.9	18.62	23.6	77.7	19.0	15	38.9	16.62	22.5	69.5	5.3
Calcium (Ca)	21	230721.7	9753.74	211039.2	247784.0	-14.1	21	247556.5	15332.50	220230.0	276971.6	-7.8
Cadmium (Cd)	16	8.2	8.20	0.0	22.9	83.9	15	10.7	21.03	0.0	82.7	139.6
Chromium (Cr)	27	49.7	26.48	17.3	110.9	4.6	27	58.4	25.40	23.8	110.2	22.8
Copper (Cu)	27	103.4	78.84	30.2	271.2	38.3	27	117.1	84.00	17.6	319.4	56.6
Iron (Fe)	24	260408.6	14308.43	233275.2	284728.0	15.9	24	246941.2	17821.97	210530.0	273850.4	9.9
Magnesium (Mg)	27	6787.9	634.83	5096.2	7512.0	39.7	27	6586.0	572.12	5121.9	7399.4	35.5
Manganese (Mn)	27	2544.0	175.72	2276.7	3005.5	17.1	27	2424.3	168.58	2160.8	2839.4	11.6
Nickel (Ni)	12	16.8	9.17	3.5	32.1	8.0	14	21.3	12.16	0.1	43.9	37.1
Zinc (Zn)	24	589.5	208.77	358.1	1053.9	31.7	24	589.0	188.00	436.6	1059.1	31.5

Note: T2 and T6, treatment systems with ochre pellets combined with floating *P. australis*; T4 and T8, treatment systems with ochre pellets only; n, number of tested samples; SD, standard deviation; Min, minimum; Max, maximum.

Table 7.6. (Continued).

(b) 7–day contact time

HC–SGW

Element	**Cement–ochre pellets in T10**						**Cement–ochre pellets in T12**					
	n	Mean	SD	Min	Max	Accumulation (%)	n	Mean	SD	Min	Max	Accumulation (%)
Aluminium (Al)	18	14775.3	1203.45	12561.2	16514.0	7.9	18	16130.6	969.31	14621.8	17984.0	17.8
Boron (B)	15	97.3	15.87	75.1	127.5	163.5	15	97.4	15.84	79.2	124.4	163.7
Calcium (Ca)	21	210908.8	15683.93	182122.4	234072.0	-21.5	21	242023.8	11280.09	217711.6	258784.8	-9.9
Cadmium (Cd)	12	152.1	42.83	65.7	203.7	3306.4	12	265.1	50.32	169.8	356.4	5838.1
Chromium (Cr)	27	88.7	38.06	42.7	193.4	86.6	27	95.8	34.21	51.5	197.2	101.5
Copper (Cu)	27	133.3	76.88	8.2	335.7	78.22	27	130.5	80.23	42.4	329.5	74.5
Iron (Fe)	24	246726.8	14898.61	216570.8	269527.6	9.8	24	243388.2	16023.00	210822.0	265205.2	8.4
Magnesium (Mg)	27	7898.1	751.94	5926.2	8885.0	62.5	27	8194.8	712.29	6240.2	8976.9	68.7
Manganese (Mn)	27	2508.1	172.89	2249.0	2944.5	15.5	27	2522.1	255.90	2104.0	3050.9	16.1
Nickel (Ni)	18	26.4	19.06	1.4	66.8	69.9	20	30.7	20.60	2.3	73.2	97.8
Zinc (Zn)	24	577.2	165.32	421.4	1005.7	28.9	24	626.7	189.38	441.2	1032.5	40.0

LC–SGW

Element	**Cement–ochre pellets in T14**						**Cement–ochre pellets in T16**					
	n	Mean	SD	Min	Max	Accumulation (%)	n	Mean	SD	Min	Max	Accumulation (%)
Aluminium (Al)	18	16492.5	903.51	15021.5	17988.1	20.4	18	16874.5	914.04	15309.2	18198.2	23.2
Boron (B)	15	52.9	17.21	33.3	83.2	43.3	15	53.1	17.16	35.4	82.3	43.9
Calcium (Ca)	21	229105.5	11801.71	208161.6	251568.0	-14.7	21	242354.2	9066.79	224942.0	256550.4	-9.7
Cadmium (Cd)	18	80.4	115.69	0.0	298.6	1701.8	18	11.8	10.14	0.0	26.2	164.3
Chromium (Cr)	27	139.9	95.20	25.8	325.5	194.3	27	68.1	33.26	29.5	153.0	43.2
Copper (Cu)	27	114.8	75.64	35.3	282.2	53.5	27	106.9	78.81	40.0	288.2	42.9
Iron (Fe)	24	251949.5	16346.18	221098.8	278889.6	12.2	24	254510.0	13227.47	230163.2	274306.4	13.3
Magnesium (Mg)	27	6613.5	543.72	5177.4	7299.5	36.1	27	6492.4	511.48	5170.8	6962.6	33.6
Manganese (Mn)	27	2461.8	171.87	2239.3	2892.3	13.4	27	2529.9	144.24	2302.0	2855.0	16.5
Nickel (Ni)	11	22.2	11.61	4.9	42.2	43.2	17	29.0	20.60	0.4	59.2	86.6
Zinc (Zn)	24	576.3	187.77	402.4	1054.8	28.7	24	578.2	175.57	449.2	1013.4	29.1

Note: T2 and T6, treatment systems with ochre pellets combined with floating *P. australis*; T4 and T8, treatment systems with ochre pellets only; n, number of tested samples; SD, standard deviation; Min, minimum; Max, maximum.

The statistical analysis showed that accumulated concentrations of all considered elements in ochre pellets after the end of the treatment experiment were significantly higher, except for Ca and Ni. The Ca concentrations which were significantly lower, than those in ochre pellets before commencing the experiment. While, the accumulated Ni concentrations did not increase significantly in ochre pellets before and after treatment in almost treatment system. Additionally, in systems T6 (ochre pellets and *P. australis*) and T8 (only ochre pellets) treating LC–SGW for 2–day contact time, the accumulated B, Cd, Cr and Cu also did not increase significantly (Table 7.7).

In treatment of HC–SGW for 2–day contact time, presence of *P. australis* in combination with ochre pellets in system T2 significantly increased the concentrations of Al, B, Ca, Cr, Cu, Fe and Zn which accumulated in ochre pellets compared to the ochre pellets in system T4 (2–day; HC–SGW; only ochre pellets). However, that effect was different in the 7–day contact time of system T10 (HC–SGW; ochre pellets with floating *P. australis*), where concentrations of Al, Ca and Cd were significantly lower than those concentrations which accumulated in the ochre pellets of system T12 (7–day; HC–SGW; only ochre pellets), as shown in Table 7.8. A considerable relationship was indicated between the presence of *P. australis* and the increasing of accumulation rate of elements in ochre pellets within the same treatment system (Figure 7.18). This could be linked to the respiration of rhizomes and production of carbon dioxide and acidic exudates, coupled with production of organic acids during the biodegradation of organic matter by microorganisms (Sooknah and Wilkie, 2004; Iamchaturapatr et al., 2007; Lynch et al., 2015). Those acidic by–products cause a significant reduction in the pH value of the water, converting insoluble particle metals to dissolved form which could be adsorbed by ochre pellets faster than they could be taken up by *P. australis* due to the chemical reactions (Figures 7.18 and 7.19).

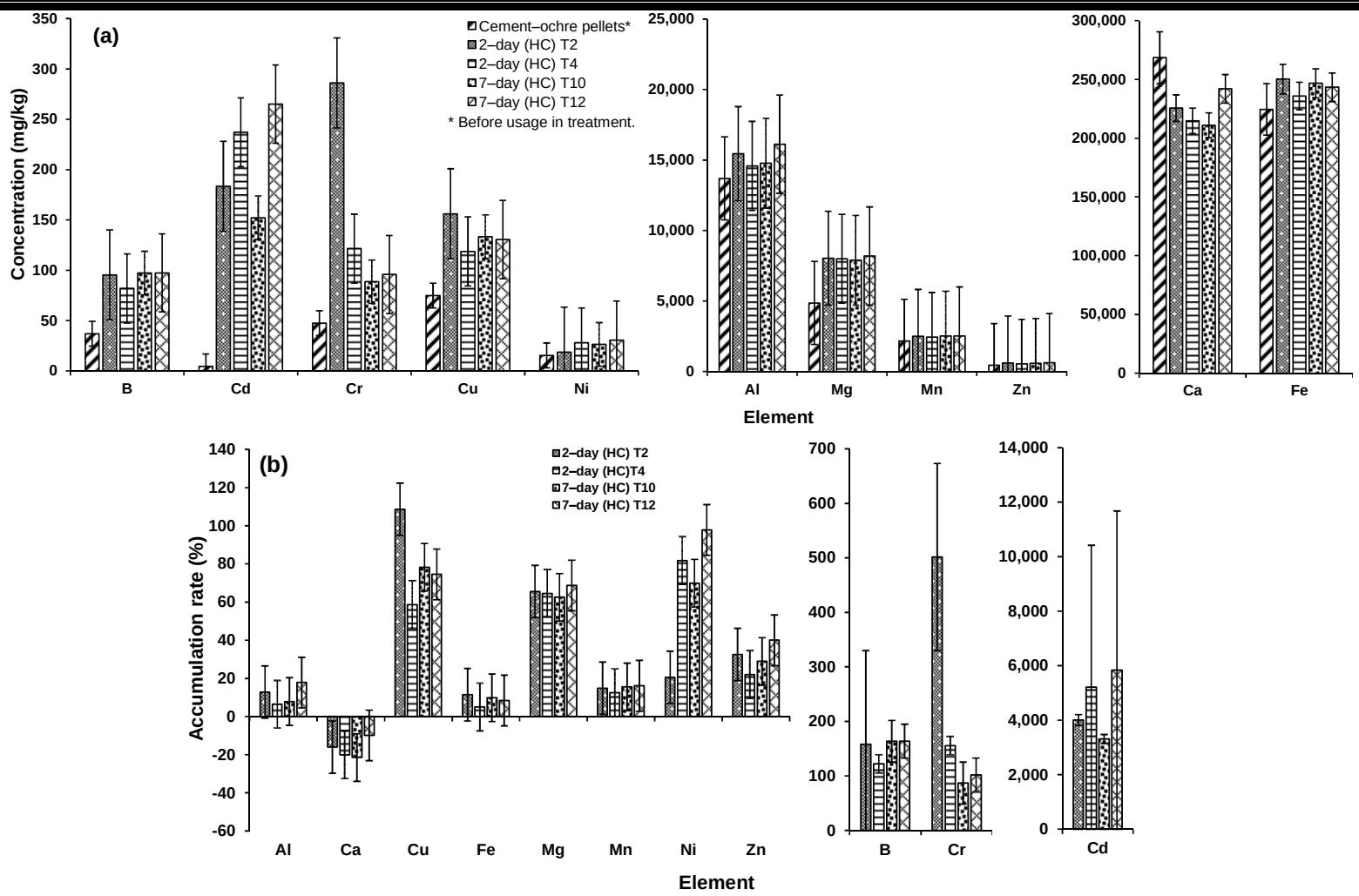


Figure 7.18. Effect of *P. australis* and treatment time on the adsorption of trace element concentrations by cement–ochre pellets treating HC–SGW in various floating treatment wetlands; a) concentrations, and b) accumulation percentage.

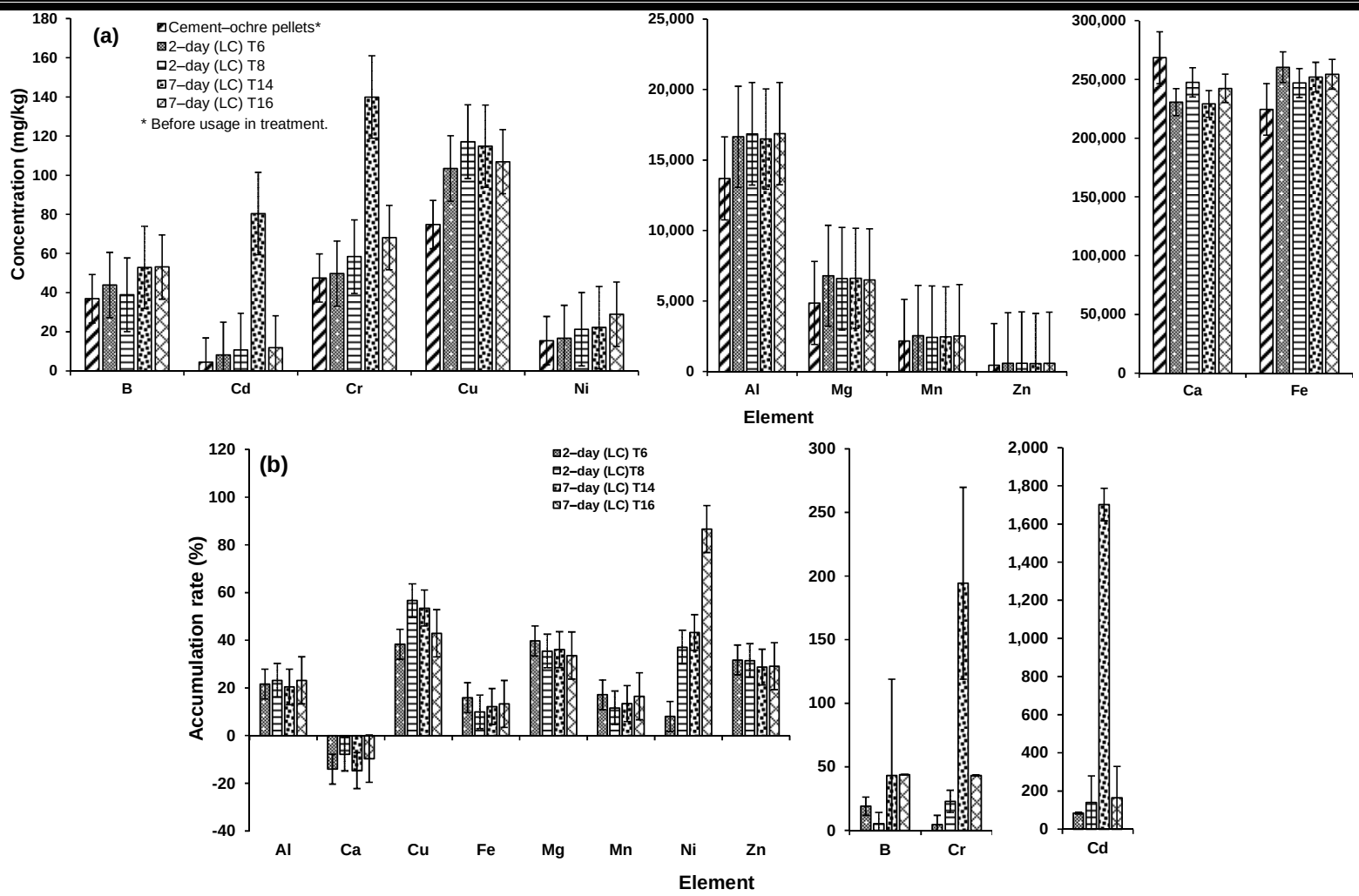


Figure 7.19. Effect of *P. australis* and treatment time on the adsorption of trace element concentrations by cement–ochre pellets treating LC–SGW in various floating treatment wetlands; a) concentrations, and b) accumulation percentage.

Table 7.7. The significant differences in comparisons of accumulated concentrations (mg/kg) of trace elements in ochre pellets before and after treating synthetic greywater (SGW) in systems (T).

HC–SGW

Concentrations in ochre pellets before treatment with:

	2–day contact time						**7–day contact time**					
	T2			**T4**			**T10**			**T12**		
Element	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
Aluminium	0.131	T–test	<0.001	0.546	T–test	0.009	0.252	T–test	0.003	0.386	T–test	<0.001
Boron	0.102	T–test	<0.001	0.290	T–test	<0.001	0.071	T–test	<0.001	0.053	T–test	<0.001
Calcium	0.123	T–test	<0.001	0.016	M–W	<0.001	0.019	M–W	<0.001	0.908	T–test	<0.001
Cadmium	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Chromium	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	0.002	M–W	<0.001
Copper	<0.001	M–W	<0.001	<0.001	M–W	0.003	<0.001	M–W	0.001	<0.001	M–W	0.002
Iron	0.136	T–test	<0.001	0.374	T–test	0.013	0.324	T–test	<0.001	0.062	T–test	<0.001
Magnesium	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Manganese	0.062	T–test	<0.001	0.011	M–W	<0.001	0.190	T–test	<0.001	0.003	M–W	<0.001
Nickel	0.393	T–test	0.441	0.006	M–W	0.045	0.045	M–W	0.103	0.023	M–W	0.018
Zinc	0.003	M–W	<0.001	<0.001	M–W	0.002	<0.001	M–W	<0.001	<0.001	M–W	<0.001

LC–SGW

Concentrations in ochre pellets before treatment with:

	2–day contact time						**7–day contact time**					
	T6			**T8**			**T14**			**T16**		
Element	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
Aluminium	0.042	M–W	<0.001	0.089	T–test	<0.001	0.103	T–test	<0.001	0.022	M–W	<0.001
Boron	0.057	T–test	0.281	0.014	M–W	0.539	0.223	T–test	0.014	0.116	T–test	0.013
Calcium	0.119	T–test	<0.001	0.771	T–test	<0.001	0.132	T–test	<0.001	0.604	T–test	<0.001
Cadmium	<0.001	M–W	0.164	<0.001	M–W	0.556	<0.001	M–W	0.006	<0.001	M–W	0.047
Chromium	<0.001	M–W	0.710	<0.001	M–W	0.082	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Copper	<0.001	M–W	0.056	<0.001	M–W	0.019	<0.001	M–W	0.006	<0.001	M–W	0.014
Iron	0.111	T–test	<0.001	0.041	M–W	<0.001	0.142	T–test	<0.001	0.087	T–test	<0.001
Magnesium	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Manganese	0.134	T–test	<0.001	0.190	T–test	<0.001	0.193	T–test	<0.001	0.140	T–test	<0.001
Nickel	0.275	T–test	0.742	0.605	T–test	0.166	0.449	T–test	0.124	0.027	M–W	0.073
Zinc	<0.001	M–W	0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001

Note: HC, high concentration; LC, low concentration; SGW, synthetic greywater; T2, T6, T10, and T14, treatment systems with floating *P. australis* and ochre pellets; T4, T8, T12, and T16, treatment systems with ochre pellets only; Shapiro–Wilk (check for normality), normally distributed data if $p > 0.05$ using T–test and non–normally distributed data if $p < 0.05$ using Mann–Whitney U–test; p value, significantly different if $p < 0.05$ and not significantly different if $p > 0.05$; M–W, Mann–Whitney U–test.

Table 7.8. Significant values of the statistical analysis for digested ochre pellet samples to investigate (a) the effects of *P. australis* on trace element accumulations (mg/kg) into ochre pellets, and (b) the effects of increasing contact time and pollutant loads on trace element accumulations (mg/kg) into ochre pellets combined with *P. australis* in treatment systems (T).

(a) Effects of *P. australis* on trace element accumulations into ochre pellets in treatment systems (T)

	2–day contact time						*7–day contact time*					
	HC–SGW (T2 & T4)			**LC–SGW (T6 & T8)**			**HC–SGW (T10 & T12)**			**LC–SGW (T14 & T16)**		
Element	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
Aluminium	0.980	T–test	0.037	0.895	T–test	0.598	0.914	T–test	0.001	0.145	T–test	0.216
Boron	0.880	T–test	0.043	0.002	M–W	0.419	0.047	M–W	0.820	0.003	M–W	0.983
Calcium	0.956	T–test	0.010	0.208	T–test	<0.001	0.198	T–test	<0.001	0.384	T–test	<0.001
Cadmium	0.062	T–test	0.090	<0.001	M–W	0.511	0.796	T–test	<0.001	<0.001	M–W	0.168
Chromium	<0.001	M–W	0.014	<0.001	M–W	0.229	0.001	M–W	0.257	<0.001	M–W	0.009
Copper	<0.001	M–W	0.014	<0.001	M–W	0.473	<0.001	M–W	0.421	<0.001	M–W	0.431
Iron	0.511	T–test	0.008	0.185	T–test	0.06	0.156	T–test	0.459	0..549	T–test	0.554
Magnesium	<0.001	M–W	0.856	<0.001	M–W	0.122	<0.001	M–W	0.084	<0.001	M–W	0.164
Manganese	0.759	T–test	0.391	0.338	T–test	0.014	0.908	T–test	0.815	0.079	T–test	0.121
Nickel	0.009	M–W	0.166	0.902	T–test	0.303	0.054	T–test	0.507	0.250	T–test	0.279
Zinc	<0.001	M–W	0.022	<0.001	M–W	0.496	<0.001	M–W	0.083	<0.001	M–W	0.635

(b) Effects of contact time and pollutant loads on trace element accumulations into ochre pellets in treatment systems (T)

	Effect of contact time						*Effect of pollutant loads*					
	HC–SGW (T2 & T10)			**LC–SGW (T6 & T14)**			**2–day contact time (T2 & T6)**			**7–day contact time (T10 & T14)**		
Element	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
Aluminium	0.966	T–test	0.103	0.946	T–test	0.673	0.842	T–test	0.008	0.604	T–test	<0.001
Boron	0.095	T–test	0.739	0.028	M–W	0.101	0.087	T–test	<0.001	0.174	T–test	<0.001
Calcium	0.688	T–test	0.002	0.899	T–test	0.631	0.656	T–test	0.152	0.525	T–test	<0.001
Cadmium	0.181	T–test	0.153	<0.001	M–W	0.038	0.001	M–W	<0.001	0.002	M–W	0.018
Chromium	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	0.144
Copper	<0.001	M–W	0.223	<0.001	M–W	0.243	<0.001	M–W	0.005	<0.001	M–W	0.110
Iron	0.434	T–test	0.445	0.499	T–test	0.063	0.502	T–test	0.030	0.688	T–test	0.253
Magnesium	0.003	M–W	0.355	<0.001	M–W	0.154	0.476	T–test	<0.001	0.118	T–test	<0.001
Manganese	0.502	T–test	0.778	0.054	T–test	0.088	0.517	T–test	0.326	0.034	M–W	0.250
Nickel	0.017	M–W	0.253	0.451	T–test	0.222	0.189	T–test	0.649	0.064	T–test	0.522
Zinc	<0.001	M–W	0.194	<0.001	M–W	0.967	<0.001	M–W	0.167	<0.001	M–W	0.741

Note: HC, high concentration; LC, low concentration; SGW, synthetic greywater; T2, T6, T10, and T14, treatment systems with floating *P. australis* and ochre pellets; T4, T8, T12, and T16, treatment systems with ochre pellets only; Shapiro–Wilk (check for normality), normally distributed data if $p > 0.05$ using T–test and non–normally distributed data if $p < 0.05$ using Mann–Whitney U–test; p value, significantly different if $p < 0.05$ and not significantly different if $p > 0.05$; M–W, Mann–Whitney U–test.

In treatment of LC–SGW for 2–day contact time in system T6 (combination of ochre pellets with floating *P. australis*), the presence of floating *P. australis* significantly affected the decrease in Ca and increase in Mn contents which accumulated in the ochre pellets compared with those contents in the ochre pellets of system T8 (2–day; LC–SGW; only ochre pellets), as shown in Figure 7.19. In treatment of LC–SGW for 7–day contact time, the presence of *P. australis* significantly affected the decrease in Ca, and increase in Cd and Cr contents in the ochre pellets of system T14 (combination of ochre pellets with floating *P. australis*) compared with those contents in the ochre pellets of system T16 (2–day; LC–SGW; only ochre pellets), as shown in Tables 7.4 and 7.6. It is expected that significant decreases in Ca contents of ochre pellets is the main reason for significant increasing in both pH value and Ca concentrations in the outflow SGW of systems containing ochre pellets (Heal et al., 2003, Littler et al., 2013).

In addition, increasing contact time of treatment of HC–SGW in system T10 led to decrease the accumulated contents of Ca and Cr in the ochre pellets of system T10 compared to those accumulated concentrations in the ochre pellets of system T2. In treatment of LC–SGW, increasing contact time significantly affected the increase in concentrations of accumulated Cd and Cr in the ochre pellets of system T14 compared with those accumulated in the ochre pellets of system T6 (Tables 7.4, 7.6 and Figure 7.18 and 7.19). It is possible that ochre pellets could be fully saturated with some elements such as Cr (in system T10) which remobilised back into the water column after adsorption (Sibrell et al., 2009).

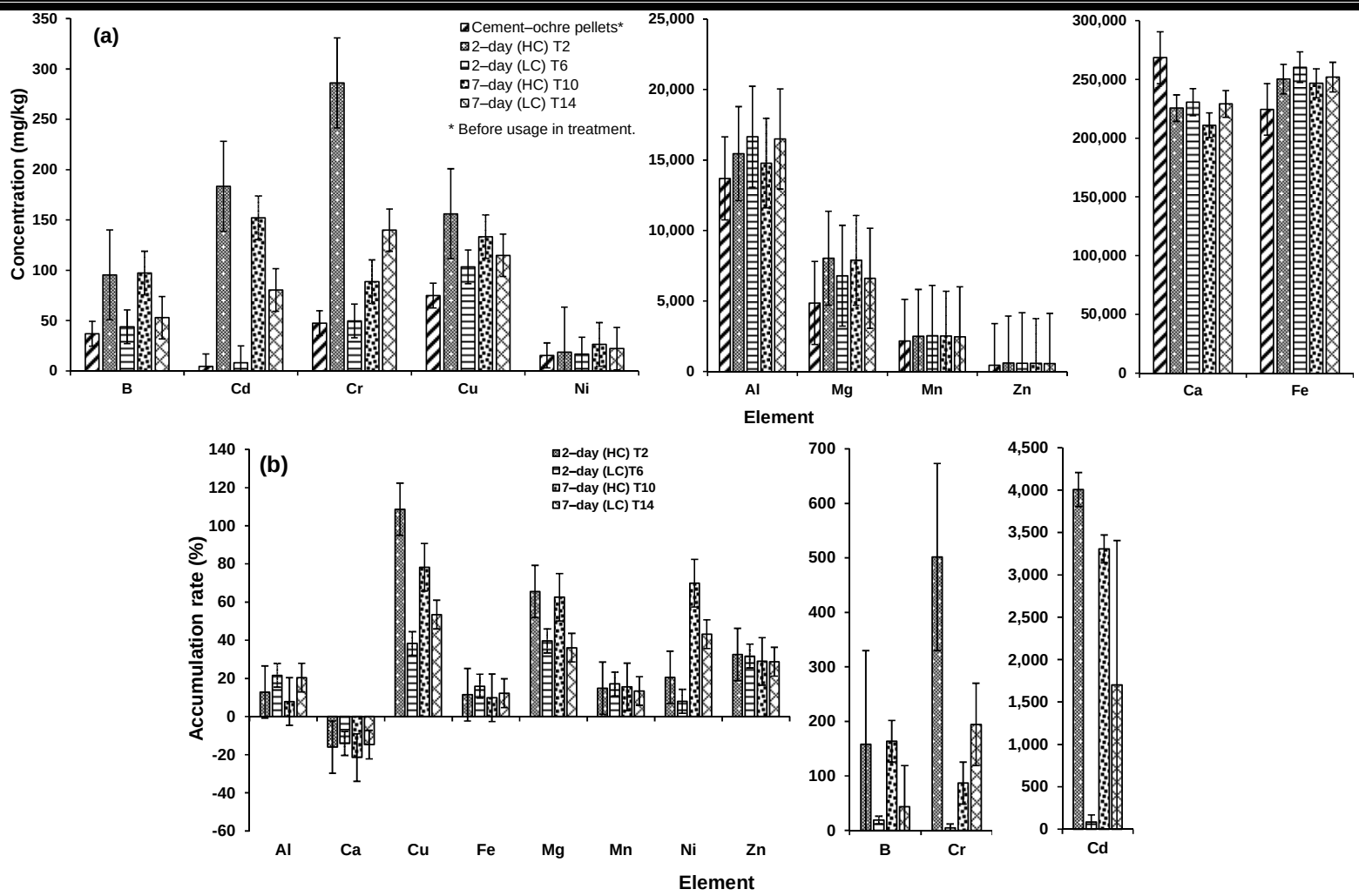


Figure 7.20. Effect of pollutant loads of SGW on the adsorption of trace element concentrations by cement–ochre pellets in various floating treatment wetlands; a) concentrations, and b) accumulation percentage.

In systems using a combination of ochre pellets and *P. australis*, the concentrations of B, Cd and Mg, (Cr and Cu in system T2 of 2–day contact time), which accumulated in ochre pellets of systems treating HC–SGW (T2: 2–day and T10: 7–day), were higher than those values in ochre pellets of systems T6 (2–day) and T14 (7–day), respectively, which were treating LC–SGW. However, contents of Al and Fe (T6: 2–day contact time), and Al and Ca (T14: 7–day contact time) which accumulated in the ochre pellets were higher in treatment of LC–SGW compared with corresponding systems T2 (2–day) and T10 (7–day), respectively, which were treating HC–SGW (Table 7.6 and Figure 7.20).

In observation of the amount of adsorbed elements, there was significant correlation with the corresponding initial trace element concentrations of greywater. Therefore, a high remediation of an element at low concentration of greywater indicates sufficient availability of vacant places on or within ochre pellets to sustain the adsorbed metals (Sahoo et al., 2014).

7.4. Trace Element Accumulated in Tissues of *P. australis*

The averages of trace element concentrations accumulated in overall tissues of *P. australis* (Table 7.7) were statistically compared to those accumulated concentrations in *P. australis* which floated in control wetlands (C), and wetlands receiving clean tap water (TW), to investigate the significant performance of each wetland (Table 7.9).

The statistical analysis showed that all considered elements, except Na which had significantly low accumulation, had significantly higher accumulation in *P. australis* of all treatment systems (T) than those in control systems (C). However, in treatment of SGW in wetlands with a combination of ochre pellets and *P. australis* at 2–day contact time, Mg accumulated in *P. australis* of wetlands T2 (HC–SGW) and B, Mg and Zn in

wetland T6 (LC–SGW) were significantly lower than those concentrations in *P. australis* of wetland C1 (2–day; TW), while there were no significant differences in concentrations of Cu in wetland T6. In 7–day contact time, Cu, Mg, Mn and Zn accumulated in *P. australis* of wetland T14 (LC–SGW) were significantly lower compared to those accumulated values in *P. australis* of control wetland C3 (7–day; TW), as shown in Table 7.10 and Figure 7.21.

In wetlands treating SGW for 7–day contact time with only floating *P. australis*, Ca accumulated in *P. australis* of wetland T9 (HC–SGW), and Ca and Mg accumulated in *P. australis* of wetlands T13 (LC–SGW) were less than those concentrations accumulated in *P. australis* of control wetland C3. Furthermore, the statistical analysis revealed that presence of ochre pellets in combination with *P. australis* in wetlands treating SGW significantly affected the reduction of the concentrations of all considered trace elements which accumulated in *P. australis* tissues, except for Ca which increased significantly, compared to those values accumulated in *P. australis* tissues in wetlands treating both types of SGW with only *P. australis* at both contact times of treatment (Table 7.9 and Figure 7.21). However, B and Cr accumulated in *P. australis* of wetland T2 (2–day; HC–SGW; combination of ochre pellets with *P. australis*), and Na accumulated in *P. australis* of wetland T14 (7–day; LC–SGW; combination of ochre pellets with *P. australis*) were significantly higher than those accumulated values in *P. australis* of wetlands T1 (2–day; HC–SGW: only *P. australis*) and T13 (7–day; LC–SGW; only *P. australis*), respectively (Table 7.11). This refers to the contribution of ochre pellets in adsorption of trace elements, however the increasing in Ca concentration was linked to the presence of ochre pellets in the treatment systems (Heal et al., 2003).

Table 7.9. The average concentrations (mg/kg) of detected trace elements which accumulated in *P. australis* at the end of the experiment period for treatment in systems (T); (a) high pollutant concentrations synthetic greywater (HC–SGW), (b) low pollutant concentrations synthetic greywater (LC–SGW) and (c) control wetlands receiving tap water (TW).

a) HC–SGW

	2–day treatment contact time										*7–day treatment contact time*									
	Dry mass:	***507.7 g***					***734.7 g***					***765.5 g***					***604.8 g***			
Element	***P. australis* in T1**					***P. australis* in T2**					***P. australis* in T9**					***P. australis* in T10**				
	n	Mean	SD	Min	Max	n	Mean	SD	Min	Max	n	Mean	SD	Min	Max	n	Mean	SD	Min	Max
Aluminium (Al)	24	1421.9	249.13	1194.6	1873.8	24	245.6	73.37	128.0	389.4	24	530.1	69.76	422.0	700.9	24	284.2	68.23	228.0	520.3
Boron (B)	15	15.1	4.00	10.3	24.5	15	18.2	3.42	12.5	24.6	15	14.8	3.40	10.7	21.1	15	8.1	3.33	4.6	13.9
Calcium (Ca)	27	4001.2	432.00	3356.4	4856.4	27	8915.8	1363.03	6362.2	10957.0	27	3996.0	214.81	3592.5	4450.6	27	6888.2	551.98	5978.8	8015.4
Cadmium (Cd)	18	1782.0	321.21	1301.7	2392.5	18	494.6	136.49	282.4	656.4	18	1293.2	146.24	1000.4	1541.1	18	467.1	113.89	316.5	663.5
Chromium (Cr)	27	1718.0	266.73	1389.0	2135.0	27	248.1	71.50	140.8	312.4	27	870.1	66.43	747.5	968.4	27	193.7	24.56	157.3	226.4
Copper (Cu)	27	477.2	74.23	394.3	597.5	27	106.2	29.60	62.1	137.3	27	347.0	28.27	302.3	398.1	27	101.3	14.07	83.6	126.4
Iron (Fe)	21	2799.4	418.29	2408.0	3414.4	21	508.7	144.41	296.4	639.5	21	2238.2	163.12	1997.1	2451.8	21	662.9	45.37	592.9	722.8
Magnesium (Mg)	27	1171.3	94.90	1020.9	1320.5	27	871.9	71.90	744.7	947.3	27	1284.6	52.18	1190.4	1382.3	27	1151.8	36.08	1077.7	1220.6
Manganese (Mn)	27	533.1	74.49	450.3	649.8	27	341.5	66.43	236.2	403.5	27	501.1	30.08	453.1	539.1	27	167.2	20.23	141.0	213.2
Sodium (Na)	12	1735.2	96.91	1602.1	1886.3	12	1787.9	62.90	1677.6	1875.6	12	1930.3	111.33	1800.7	2119.4	12	1569.4	23.23	1515.1	1591.8
Nickel (Ni)	27	37.5	14.75	12.2	91.1	27	20.8	13.31	2.2	60.5	27	90.0	12.77	61.2	130.0	27	24.8	11.34	0.0	54.8
Zinc (Zn)	24	1072.2	186.08	806.6	1467.2	24	333.4	115.66	194.6	730.4	24	699.9	72.98	551.8	872.1	24	241.9	71.35	155.8	494.5

Table 7.9. (Continued).

b) LC–SGW

	2–day treatment contact time										7–day treatment contact time									
	Dry mass:	***649.2 g***				***448.8 g***					***908.2 g***					***395.3 g***				
	***P. australis* in T5**					***P. australis* in T6**					***P. australis* in T13**					***P. australis* in T14**				
Element	n	Mean	SD	Min	Max	n	Mean	SD	Min	Max	n	Mean	SD	Min	Max	n	Mean	SD	Min	Max
Aluminium (Al)	24	744.4	56.75	682.1	861.0	24	107.8	46.81	69.4	283.4	24	207.3	54.43	144.2	355.0	24	158.6	67.16	101.26	373.3
Boron (B)	15	7.6	4.00	3.6	15.1	15	4.4	2.34	1.6	8.6	15	7.8	4.32	3.6	16.7	15	5.0	2.54	2.16	9.8
Calcium (Ca)	27	3533.9	315.71	2976.5	3988.5	27	6516.4	355.10	5961.2	7196.9	27	3390.9	226.85	3064.6	3892.6	27	10048.7	1048.10	7973.12	11941.3
Cadmium (Cd)	18	443.9	38.05	368.8	501.3	18	45.2	19.39	5.3	76.4	18	215.3	34.66	149.8	274.7	18	44.7	17.20	6.74	75.9
Chromium (Cr)	27	109.8	5.43	98.7	119.8	27	35.8	3.21	30.0	44.4	27	51.6	11.22	38.4	72.5	27	22.5	4.23	16.80	32.7
Copper (Cu)	27	259.0	9.49	238.3	277.9	27	25.4	3.23	19.9	33.7	27	93.6	8.74	80.7	110.3	27	23.9	7.06	13.47	53.6
Iron (Fe)	21	1003.8	198.84	787.4	1285.0	21	307.0	9.68	284.8	325.7	21	466.8	67.52	398.5	569.0	21	268.6	10.14	247.01	284.8
Magnesium (Mg)	27	1089.7	59.19	1008.1	1204.5	27	468.4	15.23	432.4	488.5	27	1028.7	47.19	953.7	1125.0	27	452.8	29.91	391.18	488.2
Manganese (Mn)	27	335.8	50.54	260.1	392.7	27	64.3	11.36	47.8	107.9	27	257.2	21.88	235.9	294.2	27	63.3	9.58	53.87	89.8
Sodium (Na)	12	1069.9	39.44	983.1	1141.7	12	905.5	33.77	847.4	962.4	12	1192.6	26.15	1156.1	1232.1	12	1453.2	66.54	1340.67	1557.8
Nickel (Ni)	27	57.6	14.59	26.4	95.9	27	12.6	8.69	5.4	48.9	27	22.1	10.37	1.1	59.9	27	12.7	7.85	3.61	33.2
Zinc (Zn)	24	715.1	84.90	582.5	945.7	24	94.7	37.46	62.4	214.6	24	265.9	60.60	180.4	436.6	24	109.6	59.34	57.29	275.2

c) Roots and rhizomes analysis

	Dry mass:	***856.3 g***				***567.8 g***				
	2–day contact time (*P. australis* in C1)					**7–day contact time (*P. australis* in C3)**				
Element	n	Mean	SD	Min	Max	n	Mean	SD	Min	Max
Aluminium (Al)	24	14.7	15.74	0.0	55.7	24	51.3	17.46	23.9	91.7
Boron (B)	15	7.2	3.47	4.4	14.6	15	6.3	3.63	3.0	13.6
Calcium (Ca)	27	3468.5	222.86	3041.0	3920.0	27	4284.6	267.06	3785.0	4831.2
Cadmium (Cd)	18	16.7	16.66	0.0	48.3	18	23.1	17.68	0.0	65.8
Chromium (Cr)	27	9.1	6.18	1.3	24.5	27	18.0	2.88	14.3	26.7
Copper (Cu)	27	29.3	16.01	13.6	54.7	27	30.5	3.65	25.8	39.4
Iron (Fe)	21	122.5	12.08	102.0	146.9	21	230.9	16.98	208.9	266.9
Magnesium (Mg)	27	1092.7	61.33	971.6	1191.9	27	1185.1	82.97	1061.3	1337.6
Manganese (Mn)	27	44.1	9.72	35.8	77.8	27	82.5	9.43	72.5	119.6
Sodium (Na)	12	2969.3	177.33	2717.0	3280.8	12	2139.8	202.84	1939.8	2567.3
Nickel (Ni)	27	9.2	13.58	0.2	66.7	27	12.2	15.02	0.0	63.0
Zinc (Zn)	24	221.2	46.60	165.1	342.5	24	251.5	53.76	168.3	353.0

Note: HC, high concentration; LC, low concentration; SGW, synthetic greywater; T1, T5, T9 and T13, treatment systems with only floating *P. australis*; T2, T6, T10 and T14, treatment systems with floating *P. australis* and ochre pellets; C1, control wetland contains floating *P. australis* in tap water with 2–day contact time; C3, control wetland contains floating *P. australis* in tap water with 7–day contact time.

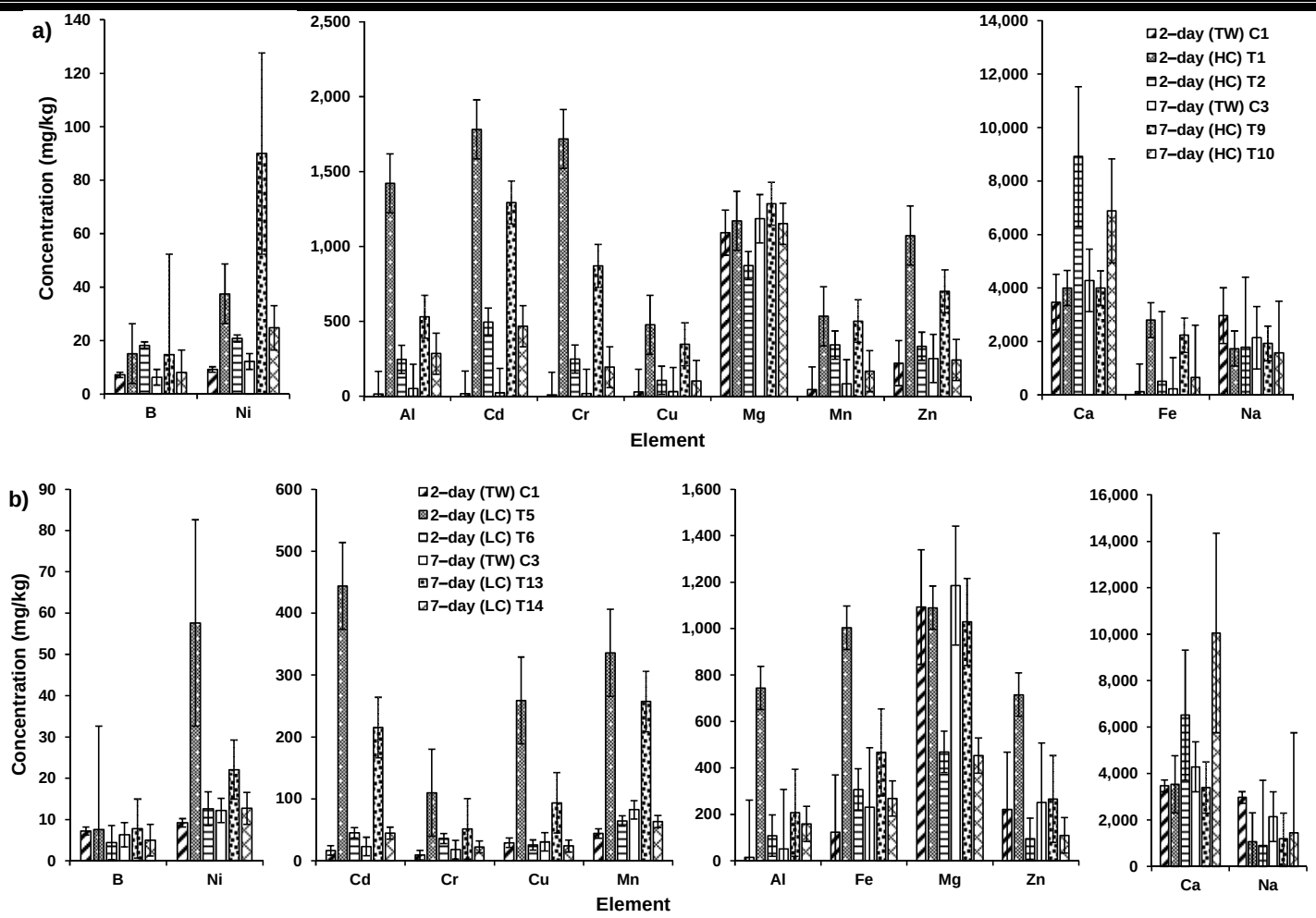


Figure 7.21. Effect of cement–ochre pellets and treatment time on the adsorption of trace element concentrations by *P. australis* treating a) HC–SGW, and b) LC–SGW in various floating treatment wetlands.

Considering the effects of contact time (Table 7.11 and Figure 7.21), the concentrations of almost all elements, which accumulated in *P. australis* tissues, were significantly lower in wetlands treating both types of SGW for 7–day contact time compared to those concentrations which accumulated in *P. australis* of wetlands at 2–day contact time of treatments. This was inverse to the behaviour exhibited in control wetlands, when comparing the trace element accumulations in *P. australis* of 7–day (C3) with those of 2–day contact time (C1). However, remarkable increases were observed with increasing treatment time in accumulation of Fe, Mg and Ni in *P. australis* of wetland T10 (HC–SGW; combination of ochre pellets with *P. australis*), and Al, Ca and Na in wetland T14 (LC–SGW; combination of ochre pellets with *P. australis*) compared to those values at 2–day treatment time wetlands T2 (HC–SGW) and T6 (LC–SGW).

The statistical analysis showed that accumulation rates in tissues of *P. australis* were higher for all considered trace elements in wetlands treating HC–SGW compared to those values in wetlands treating LC–SGW, except for the Ni accumulation rate which was higher in wetland T5 treating LC–SGW (Table 7.9 and Figure 7.22). In this context, it has been reported that FTWs are capable of coping with contaminated wastewater with high pollutant variations (Keizer–Vlek et al., 2014), especially with the presence of ochre pellets which have been proven as a good adsorbent of trace elements (Sibrell, et al., 2009; Sahoo et al., 2014; Sapsford et al., 2015).

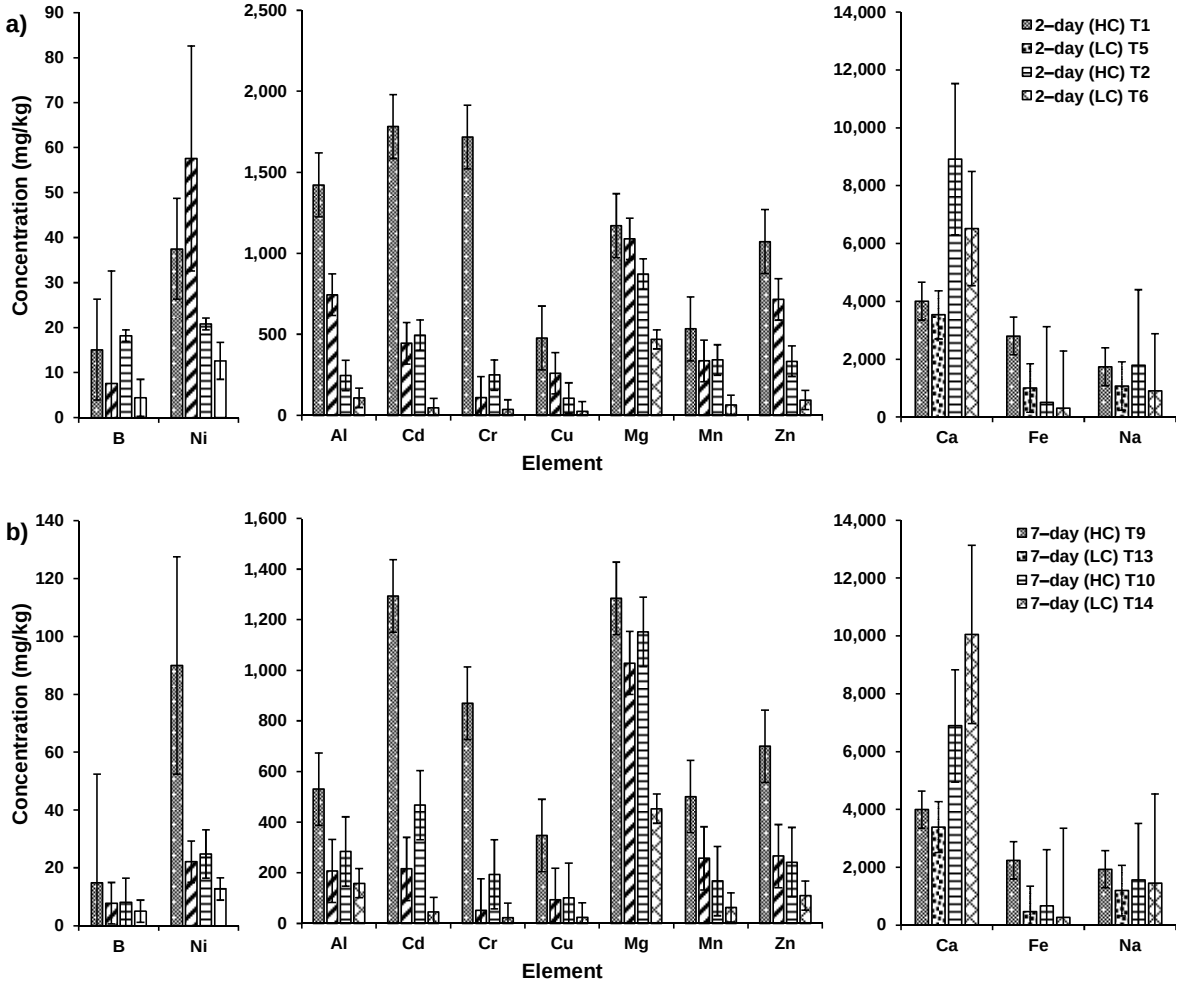


Figure 7.22. Effect of pollutant loads of SGW on the adsorption of trace element concentrations by *P. australis* in various floating treatment wetlands; a) at 2–day contact time, and b) at 7–day contact time.

Table 7.10. Significant values of the statistical analysis for accumulated trace elements (mg/kg) in *P. australis* tissue in floating treatment wetlands (T) after treating the high (HC–) and low (LC–) pollutant concentrations synthetic greywaters (SGW) compared with those elements accumulated in *P. australis* tissues of control wetlands (C); (a) 2–day contact time, and (b) 7–day contact time.

a) 2–day contact time

	HC–SGW						***LC–SGW***					
	C1 & T1			**C1 & T2**			**C1 & T5**			**C1 & T6**		
Parameter	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
Aluminium (Al)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	0.001	M–W	<0.001
Boron (B)	0.029	M–W	<0.001	0.006	M–W	<0.001	<0.001	M–W	0.917	0.001	M–W	0.003
Calcium (Ca)	0.002	M–W	<0.001	<0.001	M–W	<0.001	0.219	T–test	0.384	<0.001	M–W	<0.001
Cadmium (Cd)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	0.005	M–W	<0.001
Chromium (Cr)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Copper Cu)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	0.082
Iron (Fe)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Magnesium (Mg)	0.064	T–test	0.001	0.015	M–W	<0.001	0.103	T–test	0.853	<0.001	M–W	<0.001
Manganese (Mn)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Sodium (Na)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Nickel (Ni)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Zinc (Zn)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001

(b) 7–day contact time

	HC–SGW						***LC–SGW***					
	C3 & T9			**C3 & T10**			**C3 & T13**			**C3 & T14**		
Parameter	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
Aluminium (Al)	<0.001	M–W	<0.001	0.001	M–W	<0.001	0.009	M–W	<0.001	<0.001	M–W	<0.001
Boron (B)	0.029	M–W	<0.001	0.001	M–W	0.033	<0.001	M–W	0.152	<0.001	M–W	0.468
Calcium (Ca)	0.715	T–test	<0.001	<0.001	M–W	<0.001	0.012	M–W	<0.001	<0.001	M–W	<0.001
Cadmium (Cd)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	0.278	T–test	0.001
Chromium (Cr)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	0.004	M–W	<0.001
Copper Cu)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	0.001	M–W	<0.001
Iron (Fe)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	0.007	M–W	<0.001
Magnesium (Mg)	0.073	T–test	<0.001	<0.001	M–W	0.303	0.007	M–W	<0.001	<0.001	M–W	<0.001
Manganese (Mn)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	0.001	M–W	<0.001
Sodium (Na)	0.021	M–W	0.013	0.002	M–W	<0.001	<0.001	M–W	<0.001	0.011	M–W	<0.001
Nickel (Ni)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Zinc (Zn)	<0.001	M–W	<0.001	<0.001	M–W	0.232	0.001	M–W	0.470	0.001	M–W	<0.001

Note: HC, high concentration; LC, low concentration; SGW, synthetic greywater; T1, T5, T9 and T13, treatment systems with only floating *P. australis*; T2, T6, T10 and T14, treatment systems with floating *P. australis* and ochre pellets; C1, control wetland contains floating *P. australis* in tap water with 2–day contact time; C3, control wetland contains floating *P. australis* in tap water with 7–day contact time; Shapiro–Wilk (check for normality), normally distributed data if $p>0.05$ using T–test and non–normally distributed data if $p<0.05$ using Mann–Whitney U–test; p value, significantly different, if $p<0.05$ and not significantly different if $p>0.05$; M–W, Mann–Whitney U–test.

Table 7.11. Significant values of the statistical analysis for accumulated trace elements (mg/kg) in *P. australis* tissue in the floating treatment wetlands (T) after treating the high (HC–) and low (LC–) pollutant concentrations synthetic greywaters (SGW) to investigate: (a) effect of ochre pellets on trace element accumulation in *P. australis* tissues, (b) effect of contact time on trace element accumulation in *P. australis* tissues, and (c) effect of pollutant concentrations on trace element accumulation in *P. australis* tissues.

a) Effect of ochre pellets on trace element accumulation in *P. australis*.

	2–day contact time						***7–day contact time***					
	HC–SGW (T1 & T2)			**LC–SGW (T5 & T6)**			**HC–SGW (T9 & T10)**			**LC–SGW (T13 & T14)**		
Parameter	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
Aluminium (Al)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Boron (B)	0.264	T–test	0.027	0.001	M–W	0.015	0.073	T–test	<0.001	<0.001	M–W	0.029
Calcium (Ca)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Cadmium (Cd)	0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Chromium (Cr)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Copper Cu)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Iron (Fe)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Magnesium (Mg)	0.008	M–W	<0.001	<0.001	M–W	<0.001	0.079	T–test	<0.001	<0.001	M–W	<0.001
Manganese (Mn)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Sodium (Na)	0.187	T–test	0.128	0.043	M–W	<0.001	0.003	M–W	<0.001	0.004	M–W	<0.001
Nickel (Ni)	0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Zinc (Zn)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	0.001	M–W	<0.001

b) Effect of contact time on trace element accumulation in *P. australis*.

	HC–SGW						***LC–SGW***						***Control wetlands***			
	T1 & T9			**T2 & T10**			**T5& T13**			**T6 & T14**			**C1 & C3**			
Parameter	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	
Aluminium (Al)	<0.001	M–W	<0.001	<0.001	M–W	0.409	<0.001	M–W	<0.001	<0.001	M–W	<0.001	0.009	M–W	<0.001	
Boron (B)	0.022	M–W	0.917	0.040	M–W	<0.001	<0.001	M–W	0.724	0.002	M–W	0.290	<0.001	M–W	0.120	
Calcium (Ca)	0.290	T–test	0.956	<0.001	M–W	<0.001	0.088	T–test	0.062	<0.001	M–W	<0.001	0.084	T–test	<0.001	
Cadmium (Cd)	0.015	M–W	<0.001	0.014	M–W	0.681	<0.001	M–W	<0.001	0.137	T–test	0.933	0.004	M–W	0.194	
Chromium (Cr)	<0.001	M–W	<0.001	<0.001	M–W	0.036	<0.001	M–W	<0.001	0.004	M–W	<0.001	0.001	M–W	<0.001	
Copper Cu)	<0.001	M–W	<0.001	0.001	M–W	0.088	<0.001	M–W	<0.001	<0.001	M–W	0.008	<0.001	M–W	0.036	
Iron (Fe)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	0.029	M–W	<0.001	<0.001	M–W	<0.001	
Magnesium (Mg)	0.016	M–W	<0.001	<0.001	M–W	<0.001	0.075	T–test	<0.001	<0.001	M–W	0.095	0.047	M–W	<0.001	
Manganese (Mn)	<0.001	M–W	0.574	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	0.392	<0.001	M–W	<0.001	
Sodium (Na)	0.526	T–test	<0.001	0.007	M–W	<0.001	0.139	T–test	<0.001	<0.001	M–W	<0.001	0.020	M–W	<0.001	
Nickel (Ni)	<0.001	M–W	<0.001	<0.001	M–W	0.023	0.001	M–W	<0.001	<0.001	M–W	0.883	<0.001	M–W	0.040	
Zinc (Zn)	0.002	M–W	<0.001	<0.001	M–W	0.001	<0.001	M–W	<0.001	<0.001	M–W	0.332	0.001	M–W	0.018	

Note: HC, high concentration; LC, low concentration; SGW, synthetic greywater; T1, T5, T9 and T13, treatment systems with only floating *P. australis*; T2, T6, T10 and T14, treatment systems with floating *P. australis* and ochre pellets; C1, control wetland contains floating *P. australis* in tap water with 2–day contact time; C3, control wetland contains floating *P. australis* in tap water with 7–day contact time; Shapiro–Wilk (check for normality), normally distributed data if $p > 0.05$ using T–test and non–normally distributed data if $p < 0.05$ using Mann–Whitney U–test; p value, significantly different, if $p < 0.05$ and not significantly different if $p > 0.05$; M–W, Mann–Whitney U–test.

Table 7.11. (Continued).

c) Effect of pollutant concentrations on trace element accumulation in *P. australis*.

	2–day contact time						*7–day contact time*					
	T1 & T5			**T2 & T6**			**T9 & T13**			**T10 & T14**		
Parameter	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)	Shapiro–Wilk (p value)	Statistical test	Significance (p value)
Aluminium (Al)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	0.001	M–W	<0.001
Boron (B)	0.303	T–test	<0.001	0.006	M–W	<0.001	0.116	T–test	<0.001	0.009	M–W	0.004
Calcium (Ca)	0.065	T–test	<0.001	<0.001	M–W	<0.001	0.101	T–test	<0.001	0.001	M–W	<0.001
Cadmium (Cd)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Chromium (Cr)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Copper Cu)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Iron (Fe)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Magnesium (Mg)	0.003	M–W	0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Manganese (Mn)	0.003	M–W	0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Sodium (Na)	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	0.012	M–W	<0.001
Nickel (Ni)	0.021	M–W	<0.001	<0.001	M–W	0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001
Zinc (Zn)	0.002	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001	<0.001	M–W	<0.001

Note: HC, high concentration; LC, low concentration; SGW, synthetic greywater; T1, T5, T9 and T13, treatment systems with only floating *P. australis*; T2, T6, T10 and T14, treatment systems with floating *P. australis* and ochre pellets; C1, control wetland contains floating *P. australis* in tap water with 2–day contact time; C3, control wetland contains floating *P. australis* in tap water with 7–day contact time; Shapiro–Wilk (check for normality), normally distributed data if $p > 0.05$ using T–test and non–normally distributed data if $p < 0.05$ using Mann–Whitney U–test; p value, significantly different, if $p < 0.05$ and not significantly different if $p > 0.05$; M–W, Mann–Whitney U–test.

In this study, floating *P. australis* in a hydroponic manner has shown significant ($p < 0.05$) tolerance for remediation of the high concentrations of pollutants such as minerals and heavy elements, however, that has a negative effective on their growth rates in floating treatment wetlands.

The distribution of trace elements in the *P. australis* tissues into roots and rhizomes, stems and leaves was investigated (Table 7.12). Among the plant parts, the comparative study showed that trace elements accumulated in roots and rhizomes were significantly ($p < 0.05$) higher than those concentrations in stems and leaves (Figure 7.23), which agreed with previous research (Salt et al., 1995; Verma et al., 2016).

The accumulations of trace elements were substantially varied between stems and leaves with leaf concentrations being often higher, however, this depends on the plant uptake capacity and intracellular transportation in the plant. The leaf vacuoles are generally considered to be the main storage locations for heavy metals in plants and sequestration of heavy metals in the vacuole is also part of the tolerance mechanism (Vymazal, 2016). An exception to this is the accumulative Zn which was present in higher concentrations in stems (Table 7.12) due to the presence of growth hormone indole acetic acid, which primarily performs its function in stems (Vymazal and Brezinová; 2016).

In comparison with control wetlands, the accumulated trace elements in rhizomes of *P. australis* grown in all treatment systems were significantly ($p < 0.05$) higher in terms of Al, B, Ca, Cd, Cr, Cu, Fe, Mg, Mn and Ni. However, the concentrations of Na and Zn were indicated to be lower than those concentrations of control wetlands (Figure 7.23). The concentrations of almost all trace elements in stems and leaves were significantly ($p < 0.05$) higher in treatment systems than in control wetlands, however, accumulations of Cr, Mg, Na and Ni were lower in stems of *P. australis* in almost all treatment wetlands.

The accumulation of elements in floating *P. australis* of systems treating HC–SGW were significantly ($p < 0.05$) higher than those values in treatment of LC–SGW, through different locations of the plants, as shown by comparing Figure 7.23–a with b and Figure 7.23–c with d.

Presence of cement–ochre pellets in treatment systems had a significant ($p < 0.05$) effect on the decrease in the accumulation of trace elements in rhizomes of plants, with the exception of Ca which had significantly ($p < 0.05$) higher accumulation in *P. australis* rhizomes with the presence of ochre pellets.

Increasing contact time of treatment using a combination of ochre pellets with *P. australis* significantly ($p < 0.05$) increased accumulated trace elements in rhizomes of *P. australis* compared to those values in systems of 2–day contact time. In contrast, the accumulation in rhizomes of plants in systems of only floating macrophytes were significantly ($p < 0.05$) higher in systems of 2–day treatment time compared with systems of 7–day contact time, as shown by comparing Figure 7.23–a with c and Figure 7.23–b with d.

Table 7.12. Detected trace element concentrations (mg/kg) accumulated in *P. australis* leaves, stems, and rhizomes at the end of the experiment of floating treatment wetlands (C and T) after treatment of (a) high pollutant concentrations (HC–SGW), and (b) low pollutant concentrations synthetic greywater (LC–SGW).

a) HC–SGW

2–day treatment contact time

Element	***P. australis* in C1**			***P. australis* in T1**			***P. australis* in T2**		
	Leaf	Stem	Rhizome	Leaf	Stem	Rhizome	Leaf	Stem	Rhizome
Aluminium (Al)	25.8±22.51 (48)	6.3±13.51 (24)	25.9±26.60 (24)	108.5±69.22 (49)	36.2±20.01 (24)	4155.0±736.04 (24)	116.0±107.16 (50)	262.4±86.51 (24)	394.6±133.80 (24)
Boron (B)	18.0±5.04 (30)	2.2±3.24 (15)	1.9±2.70 (15)	11.5±3.18 (30)	4.2±3.40 (15)	28.9±6.39 (15)	15.8±5.02 (30)	31.7±5.82 (15)	8.4±3.02 (15)
Calcium (Ca)	7652.9±600.16 (54)	1866.7±126.40 (27)	1126.7±273.74 (27)	3728.7±211.48 (54)	1511.5±135.69 (27)	6779.7±1040.79 (27)	9597.3±2523.86 (54)	11008.2±2351.84 (27)	7007.6±1221.72 (27)
Cadmium (Cd)	24.3±28.99 (36)	18.7±24.08 (18)	17.3±18.62 (18)	61.5±25.40 (36)	52.5±29.30 (18)	5234.3±942.07 (18)	263.0±137.19 (36)	298.7±84.93 (18)	939.0±300.40 (18)
Chromium (Cr)	8.8±6.90 (54)	12.6±10.39 (27)	9.8±6.10 (27)	35.4±6.26 (54)	17.2±9.57 (27)	5105.7±798.88 (27)	122.7±62.97 (54)	111.1±37.32 (27)	525.4±173.61 (27)
Copper (Cu)	29.5±27.39 (54)	18.4±6.81 (27)	30.5±13.97 (27)	28.0±6.91 (54)	28.1±4.30 (27)	1375.6±219.61 (27)	48.5±22.92 (54)	59.5±15.97 (27)	216.2±73.73 (27)
Iron (Fe)	152.0±42.26 (42)	42.6±6.89 (21)	211.1±44.38 (21)	274.9±92.15 (42)	114.0±20.37 (21)	8099.5±1234.60 (21)	331.2±180.1 (42)	333.9±96.55 (21)	977.0±334.01 (21)
Magnesium (Mg)	1495.4±78.17 (54)	947.6±56.96 (27)	864.3±100.65 (27)	915.8±48.28 (54)	1093.1±124.65 (27)	1525.0±173.61 (27)	1097.5±159.23 (54)	1217.1±91.22 (27)	437.0±63.11 (27)
Manganese (Mn)	41.8±18.10 (54)	18.5±11.83 (27)	74.8±13.51 (27)	35.4±10.08 (54)	24.9±7.53 (27)	1541.4±221.46 (27)	275.3±86.73 (54)	214.5±50.97 (27)	609.5±145.57 (27)
Sodium (Na)	1412.2±435.10 (24)	2899.4±237.52 (12)	4189.1±315.20 (12)	1123.2±442.61 (24)	1613.5±135.56 (12)	2039.9±260.78 (12)	1662.3±255.47 (24)	2385.1±87.18 (12)	1089.9±71.99 (12)
Nickel (Ni)	6.1±5.15 (49)	10.2±23.75	11.6±18.00 (27)	9.7±8.67 (49)	12.2±13.75 (27)	89.9±31.14 (27)	11.7±8.61 (49)	13.1±8.15 (27)	40.8±32.57 (27)
Zinc (Zn)	131.5±54.56 (48)	407.8±55.44 (24)	135.0±68.40 (24)	129.8±69.81 (48)	374.4±129.54 (24)	2711.6±448.55 (24)	181.5±96.79 (48)	346.2±86.76 (24)	505.3±252.88 (24)

7–day treatment contact time

Element	***P. australis* in C3**			***P. australis* in T9**			***P. australis* in T10**		
	Leaf	Stem	Rhizome	Leaf	Stem	Rhizome	Leaf	Stem	Rhizome
Aluminium (Al)	51.4±30.91 (48)	10.5±16.16 (24)	110.5±26.82 (24)	105.4±114.61 (51)	50.6±28.32 (24)	1450.2±124.73 (24)	104.4±147.50 (46)	18.9±21.73 (24)	714.2±69.44 (24)
Boron (B)	14.5±5.96 (30)	3.1±3.54 (15)	1.6±2.37 (15)	18.8±4.49 (30)	10.5±3.25 (15)	15.9±4.27 (15)	10.6±4.10 (30)	2.7±2.93 (15)	11.3±4.81 (15)
Calcium (Ca)	9196.9±690.76 (54)	2179.4±214.58 (27)	1709.0±141.14 (27)	6308.2±633.96 (54)	1648.2±172.03 (27)	4399.7±346.27 (27)	6039.2±504.45 (54)	1578.9±121.61 (27)	13228.0±1847.21 (27)
Cadmium (Cd)	18.2±22.12 (36)	13.5±15.04 (18)	38.5±20.82 (18)	113.9±21.99 (36)	222.5±81.33 (18)	3548.8±381.14 (18)	95.7±128.98 (36)	40.2±18.11 (18)	1209.1±201.80 (18)
Chromium (Cr)	7.1±4.16 (54)	7.7±3.51 (27)	39.1±3.71 (27)	55.3±8.16 (54)	54.2±9.51 (27)	2506.7±193.93 (27)	26.2±6.08 (54)	19.3±3.79 (27)	538.4±72.01 (27)
Copper (Cu)	22.0±7.99 (54)	29.4±7.02 (27)	42.1±10.78 (27)	38.9±9.84 (54)	37.9±8.97 (27)	967.2±78.80 (27)	23.1±11.34 (54)	17.6±6.79 (27)	261.8±32.08 (27)
Iron (Fe)	178.0±48.84 (42)	51.6±14.02 (21)	508.9±27.54 (21)	354.8±202.89 (42)	155.4±28.47 (21)	6352.9±471.40 (21)	312.4±199.93 (42)	76.9±12.72 (21)	1572.4±215.68 (21)
Magnesium (Mg)	1589.4±84.58 (54)	1171.3±113.25 (27)	852.1±85.92 (27)	1771.4±166.71 (54)	1098.6±74.38 (27)	1096.9±56.29 (27)	1105.7±83.24 (54)	756.8±48.41 (27)	1649.1±125.36 (27)
Manganese (Mn)	114.5±16.10 (54)	33.9±12.18 (27)	104.9±10.36 (27)	84.8±13.62 (54)	77.7±15.82 (27)	1348.4±86.37 (27)	54.8±11.59 (54)	30.7±12.12 (27)	417.1±61.55 (27)
Sodium (Na)	1087.8±368.86 (24)	1846.1±187.25 (12)	3130.6±376.17 (12)	1312.8±379.90 (24)	2508.7±127.92 (12)	1633.7±66.55 (12)	1011.1±382.83 (24)	1369.1±131.62 (12)	1960.0±67.56 (12)
Nickel (Ni)	6.4±4.65 (49)	10.8±18.64 (27)	18.8±30.10 (27)	14.8±9.13 (49)	17.2±13.56 (27)	239.9±33.15 (27)	7.7±5.91 (49)	12.9±19.73 (27)	56.0±21.82 (27)
Zinc (Zn)	159.3±50.96 (48)	460.4±83.21 (24)	129.2±55.66 (24)	209.9±73.11 (48)	350.1±65.48 (24)	1542.1±135.70 (24)	81.9±49.14 (48)	144.6±45.40 (24)	498.5±160.44 (24)

Table 7.12. (Continued).

b) LC–SGW

2–day treatment contact time

Element	***P. australis* in C1**			***P. australis* in T5**			***P. australis* in T6**		
	Leaf	Stem	Rhizome	Leaf	Stem	Rhizome	Leaf	Stem	Rhizome
Aluminium (Al)	25.8±22.51 (48)	6.3±13.51 (24)	25.9±26.60 (24)	47.3±51.42 (47)	9.7±12.53 (24)	2180.1±136.80 (24)	83.7±85.84 (45)	30.0±24.09 (24)	218.9±30.12 (24)
Boron (B)	18.0±5.04 (30)	2.2±3.24 (15)	1.9±2.70 (15)	10.9±5.34 (30)	3.4±3.82 (15)	9.2±4.96 (15)	10.7±4.17 (30)	2.5±2.48 (15)	2.1±2.30 (15)
Calcium (Ca)	7652.9±600.16 (54)	1866.7±126.40 (27)	1126.7±273.74 (27)	5766.2±718.72 (54)	1551.1±149.31 (27)	3856.4±661.27 (27)	7090.8±934.14 (54)	2795.8±275.16 (27)	10398.9±999.87 (27)
Cadmium (Cd)	24.3±28.99 (36)	18.7±24.08 (18)	17.3±18.62 (18)	22.8±23.30 (36)	21.0±20.06 (18)	1281.6±115.61 (18)	22.3±16.75 (36)	29.6±24.93 (18)	86.6±31.19 (18)
Chromium (Cr)	8.8±6.90 (54)	12.6±10.39 (27)	9.8±6.10 (27)	15.1±10.34 (54)	6.6±3.38 (27)	306.8±13.45 (27)	14.0±5.25 (54)	9.4±2.98 (27)	83.3±8.69 (27)
Copper (Cu)	29.5±27.39 (54)	18.4±6.81 (27)	30.5±13.97 (27)	19.4±6.10 (54)	23.7±6.95 (27)	734.9±31.63 (27)	16.0±5.87 (54)	13.6±5.35 (27)	48.9±7.81 (27)
Iron (Fe)	152.0±42.26 (42)	42.6±6.89 (21)	211.1±44.38 (21)	166.2±61.57 (42)	51.8±4.74 (21)	2850.5±612.90 (21)	194.1±82.58 (42)	105.9±18.67 (21)	697.7±64.40 (21)
Magnesium (Mg)	1495.4±78.17 (54)	947.6±56.96 (27)	864.3±100.65 (27)	1229.9±143.27 (54)	1183.2±79.25 (27)	972.7±55.30 (27)	670.0±225.44 (54)	351.0±17.77 (27)	566.3±39.11 (27)
Manganese (Mn)	41.8±18.10 (54)	18.5±11.83 (27)	74.8±13.51 (27)	28.5±16.29 (54)	19.2±12.09 (27)	963.0±150.64 (27)	53.3±18.51 (54)	32.3±13.59 (27)	113.2±11.04 (27)
Sodium (Na)	1412.2±435.10 (24)	2899.4±237.52 (12)	4189.1±315.20 (12)	880.3±286.12 (24)	1016.9±33.66 (12)	1038.6±63.04 (12)	963.8±213.59 (24)	917.8±83.62 (12)	657.7±42.51 (12)
Nickel (Ni)	6.1±5.15 (49)	10.2±23.75 (27)	11.6±18.00 (27)	5.3±3.03 (49)	13.6±20.91 (27)	154.0±35.61 (27)	6.2±6.73 (49)	8.6±10.18 (27)	22.1±18.99 (27)
Zinc (Zn)	131.5±54.56 (48)	407.8±55.44 (24)	135.0±68.40 (24)	196.3±64.09 (48)	936.9±188.97 (24)	1032.6±130.47 (24)	87.0±57.8.0 (48)	81.7±35.62 (24)	122.7±49.37 (24)

7–day treatment contact time

Element	***P. australis* in C3**			***P. australis* in T13**			***P. australis* in T14**		
	Leaf	Stem	Rhizome	Leaf	Stem	Rhizome	Leaf	Stem	Rhizome
Aluminium (Al)	51.4±30.91 (48)	10.5±16.16 (24)	110.5±26.82 (24)	67.1±98.81 (44)	14.2±18.83 (24)	529.3±65.30 (24)	88.4±139.80 (48)	12.7±18.75 (24)	359.9±49.89 (24)
Boron (B)	14.5±5.96 (30)	3.1±3.54 (15)	1.6±2.37 (15)	15.7±5.77 (30)	2.6±3.03 (15)	5.7±4.92 (15)	9.9±3.83 (30)	2.5±2.44 (15)	2.8±2.48 (15)
Calcium (Ca)	9196.9±690.76 (54)	2179.4±214.58 (27)	1709.0±141.14 (27)	6876.1±574.30 (54)	1354.8±78.18 (27)	2302.2±258.14 (27)	6696.9±573.44 (54)	2119.7±377.36 (27)	21645.9±2540.44 (27)
Cadmium (Cd)	18.2±22.12 (36)	13.5±15.04 (18)	38.5±20.82 (18)	46.4±74.70 (36)	28.1±19.00 (18)	537.9±84.72 (18)	29.3±27.60 (36)	16.3±18.02 (18)	77.3±23.34 (18)
Chromium (Cr)	7.1±4.16 (54)	7.7±3.51 (27)	39.1±3.71 (27)	5.8±3.88 (54)	9.5±4.01 (27)	139.0±33.92 (27)	15.3±8.61 (54)	7.4±4.48 (27)	41.3±4.61 (27)
Copper (Cu)	22.0±7.99 (54)	29.4±7.02 (27)	42.1±10.78 (27)	20.7±8.19 (54)	27.1±5.80 (27)	229.5±25.71 (27)	16.0±8.16 (54)	13.1±8.21 (27)	44.0±12.74 (27)
Iron (Fe)	178.0±48.84 (42)	51.6±14.02 (21)	508.9±27.54 (21)	184.1±53.55 (42)	60.5±6.66 (21)	1194.0±157.61 (21)	206.9±56.80 (42)	58.8±8.75 (21)	594.6±25.88 (21)
Magnesium (Mg)	1589.4±84.58 (54)	1171.3±113.25 (27)	852.1±85.92 (27)	1334.3±97.65 (54)	830.0±39.89 (27)	991.7±59.51 (27)	592.9±47.24 (54)	388.3±72.28 (27)	411.3±19.46 (27)
Manganese (Mn)	114.5±16.10 (54)	33.9±12.18 (27)	104.9±10.36 (27)	46.4±14.54 (54)	53.8±14.79 (27)	675.8±70.68 (27)	51.0±12.79 (54)	32.4±9.57 (27)	108.1±9.95 (27)
Sodium (Na)	1087.8±368.86 (24)	1846.1±187.25 (12)	3130.6±376.17 (12)	873.7±393.08 (24)	701.0±86.37 (12)	1627.8±101.80 (12)	952.8±400.13 (24)	1899.8±231.87 (12)	1119.3±61.48 (12)
Nickel (Ni)	6.4±4.65 (49)	10.8±18.64 (27)	18.8±30.10 (27)	6.2±5.11 (49)	12.5±21.87 (27)	47.3±16.37 (27)	7.3±7.09 (49)	7.6±9.66 (27)	23.5±14.91 (27)
Zinc (Zn)	159.3±50.96 (48)	460.4±83.21 (24)	129.2±55.66 (24)	102.2±66.30 (48)	256.1±71.19 (24)	446.3±91.84 (24)	78.2±49.81 (48)	117.8±89.31 (24)	136.6±59.78 (24)

Note: C1 and C3, control wetlands with floating *P. australis* in tap water; T1, T5, T9, and T13, treatment systems with floating *P. australis* only; T2, T6, T19, and T14, treatment systems with combined floating *P. australis* and ochre pellets.

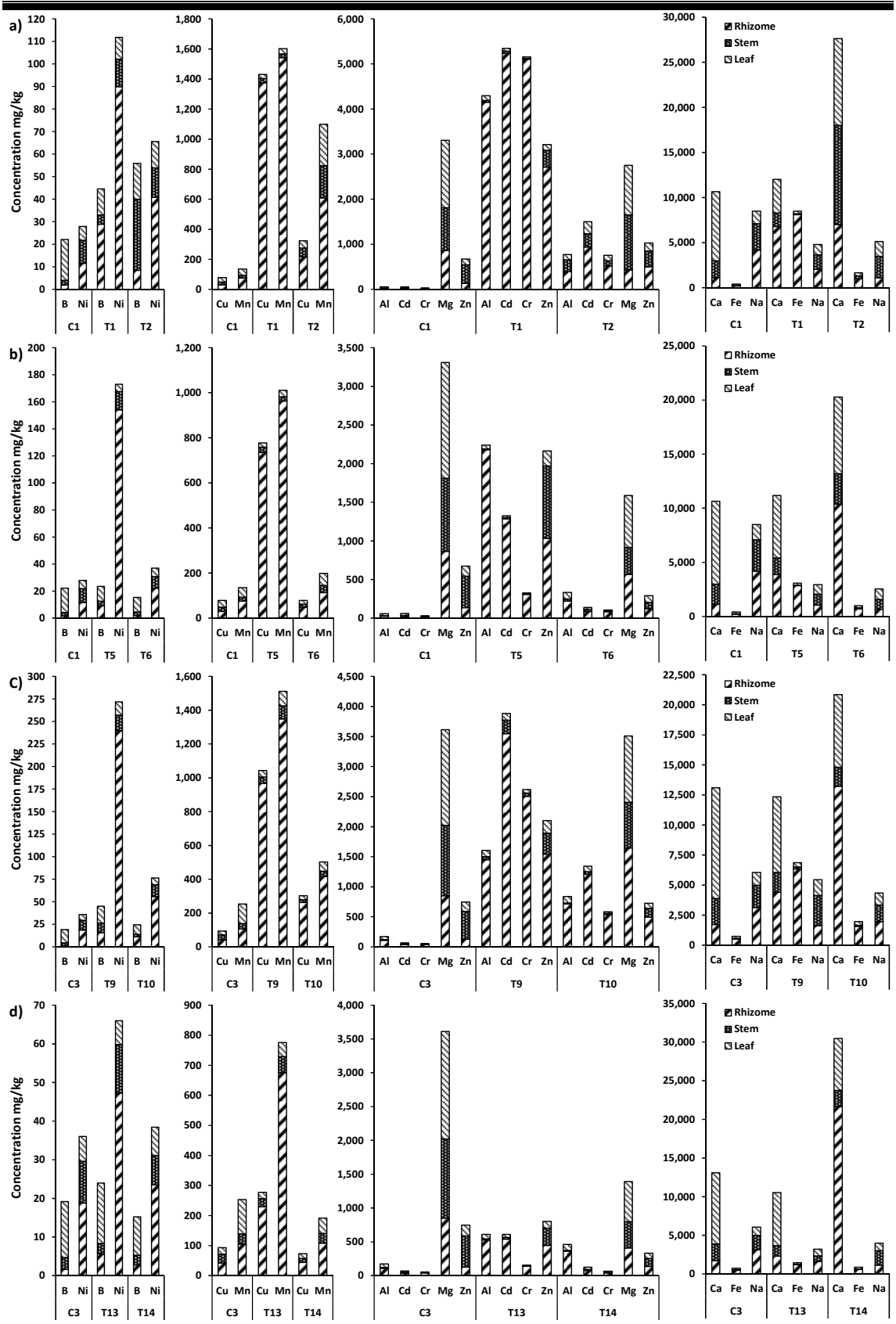


Figure 7.23. Distribution of the trace element concentrations accumulated in rhizomes, stems and leaves of *P. australis* in different designs of floating treatment wetlands.

Chapter Eight

Conclusions and Recommendations

Experimental mesocosm floating treatment wetlands of *P. australis* and cement–ochre pellets were operated and used to assess the internal processes and efficacy of the wetlands and impact of design and operational variables on treatment performance and their relationship with removal of a wide range of pollutants.

- **The main conclusions and recommendations drawn from Chapter Four are summarised as follows:**

The proposed new synthetic greywater recipes mimic real greywater well in both composition and properties. Furthermore, they provide a good matrix for microorganisms to survive and contain compounds in detectable concentrations identified as having a potentially detrimental environmental impact. The suggested recipes for LC–SGW and HC–SGW loadings are easy to prepare and replicate by others in the future. All selected materials were of chemical analytical grade. High–quantity stock solutions can be prepared and stored at 4 °C without major concern.

Throughout monitoring of the synthetic greywater properties during storage, the average values of the water quality parameters were chemically relatively stable. It has been noticed that only significant ($p < 0.05$) fluctuations in the BOD_5 for both greywater concentrations may occur. In addition, it is not recommended to store the synthetic greywater for more than two days to avoid depletion of dissolved oxygen due to development of microorganisms. Furthermore, significant changes in nitrate–nitrogen content might be noticed after two days of storage.

❖ **The main conclusions and recommendations drawn from Chapter Five are summarised as follows:**

Compared to the inflow, significant ($p < 0.05$) drops in outflow BOD_5 values were observed for all treatment systems. The presence of *P. australis* significantly ($p < 0.05$) increased the outflow BOD_5 levels compared to those of unvegetated systems, indicating that this plant contributes to pollution. No significant ($p > 0.05$) effects of vegetation on BOD_5 removals in wetlands treating different contamination loads of greywaters were recorded.

With increasing contact time, significant ($p < 0.05$) increases in BOD_5 concentrations were noted for vegetated wetlands compared with unvegetated ones. For both types of greywater, the increase of treatment time in vegetated floating wetlands can reduce the performance in BOD_5 removal, which requires biological degradation of organic matter by microorganisms.

Significant ($p < 0.05$) decreases in HC–SGW outflow COD concentrations were observed for both vegetated and unvegetated treatment systems with different contact times. After two days of treatment for LC–SGW, no significant ($p > 0.05$) differences between the COD concentrations of outflow and inflow for both vegetated and unvegetated wetlands were observed. In contrast to wetlands containing LC–SGW, the *P. australis* treating HC–SGW significantly ($p < 0.05$) impacted on reducing COD concentrations. In vegetated FTWs, higher removal rates of COD can be achieved when treating HC–SGW compared to LC–SGW. Increasing contact time significantly ($p < 0.05$) affects COD concentration reductions for both types of greywater.

The results showed that the studied treatment systems are not efficient in removing nitrogenous compounds, particularly nitrate–nitrogen, due to a lack of

biodegradable organic matter (low BOD_5 : COD ratio), which can be efficiently utilised by denitrifying bacteria. The biodegradation ratio of outflow greywater from vegetated wetlands is more promising than the corresponding ratio for unvegetated wetlands.

No significant ($p > 0.05$) effects of vegetation on the outflow NH_4–N concentrations compared were found. The treatment of low strength greywater in vegetated wetlands has shown higher removal rates for NH_4–N than the treatment of waters with high contamination. Significant ($p < 0.05$) increases in the outflow NH_4–N concentrations were noted with increasing contact time for vegetated floating treatment wetlands.

The presence of *P. australis* in floating treatment wetlands significantly ($p <0.05$) increased the outflow NO_3–N concentrations, if compared to those of unvegetated wetlands for both types of synthetic greywater. This indicates that this plant contributed to nitrogen pollution. Increasing contact time significantly ($p < 0.05$) affected the decreasing NO_3–N concentrations in the outflow of wetlands treating both types of synthetic greywater.

The TSS and turbidity values dropped significantly ($p < 0.05$) in the presence of vegetation compared with those values for unvegetated wetlands. There were no significant ($p > 0.05$) effects of the contamination load strengths on removals of TSS and turbidity from the outflow of vegetated wetlands at both contact times of treatment. The TSS and turbidity values decreased significantly ($p < 0.05$) with an increase of treatment time for the outflow of LC–SGW treating both vegetated and unvegetated wetlands. Therefore, more than 2–day contact time could improve removals of TSS, COD and NO_3–N in FTW.

Low growth rates were observed for the hydroponic plant rhizomes of *P. australis*, which might be an effect of light–induced fluorescein degradation due to the partly open water surfaces. Furthermore, the rhizomes of macrophytes are suspended in the water, and are therefore easily subject to damage, consequently affecting the removal performances. So, any buoyant mass is strongly recommended to protect floating plants. It follows that the growth of *P. australis* without soil is challenging.

Limited stand biomass and growth rate of *P. australis* could also be due to high pH values, which are known to inhibit photosynthesis causing a reduction in the macrophyte growth rate, coupled with low temperature conditions and greywater unbalanced nutrients. Therefore, dosing with secondary effluents or organic free phosphate fertilisers could improve the growth of plants. The assessment of alternative plant species is also recommended to optimise the removal performance in FTWs. The findings and recommendations of this study will help to optimise greywater treatment systems with floating wetlands by improving the nutrient balance.

❖ **The main conclusions and recommendations drawn from Chapter Six are summarised as follows:**

- Deerplay ochre sludge and its corresponding pellets are based on non–hazardous materials, and can be used in aquatic treatment systems to mobilise toxic elements.
- Cement–ochre pellets are robust and durable, and have a relatively high adsorption capacity for substances such as phosphorus.
- Calcium phosphate precipitation was a minor removal process in phosphorus treatment with cement–ochre pellets.
- Mobilisation of Fe and Al minerals from ochre pellets due to a decrease in redox potential allows for PO_4–P to take vacant places within ochre pellets.

Correspondingly, PO_4–P adsorption from greywater could occur during the movement of Ca and Fe to ochre pellet surfaces as a result of saturation. This can also lead to an increase in pH.

- Ochre–cement pellets are efficient in adsorbing and sustaining accumulated minerals and trace elements from contaminated waters.
- Significant accumulations of Cd, Cr, B, Ni, Mg, Cu, Zn and Mn indicate remediation of SGW for different treatment systems containing ochre pellets during the early stages of treatment. After successive times of treatment, ochre pellets reached their adsorption capacity in terms of Cd, Cr, Ni, Cu and Zn, but not for B, Mg and Mn with respect to the treatment of HC–SGW.
- An increase in contact time allows for the mobilisation of accumulated Ca, Cu, Mg and Zn during the treatment of LC–SGW.
- Slow adsorption rates of trace elements were linked to increasing contact times for Al, B, Ca and Zn regarding the treatment of HC–SGW, and B and Mn concerning the treatment of LC–SGW.
- The amount of adsorbed elements correlated with initial trace element concentrations in greywater. A high remediation of an element at low concentrations of greywater indicates the availability of vacant places within ochre pellets to sustain the corresponding adsorbed element, which could be B, Cd, Cr, Mg, or Zn in this study.

❖ **The main conclusions drawn from Chapter Seven are summarised as follows:**

- The presence of ochre pellets increased pH and EC of the SGW significantly ($p < 0.05$) in FTWs.
- *P. australis* neutralised pH of SGW in FTWs without ochre pellets.

- *P. australis* had no significant ($p > 0.05$) effect on pH, Eh and EC when presented with ochre pellets in FTWs.
- Increasing contact time significantly ($p < 0.05$) increased the pH in systems treating HC–SGW with a combination of ochre pellets and *P. australis*.
- The presence of ochre pellets in combination with *P. australis* in the system significantly ($p < 0.05$) increased the turbidity, TSS, and colour.
- The presence of *P. australis* in combination with ochre pellets in treatment system T10 decreased the turbidity, TSS and colour significantly ($p < 0.05$).
- TSS and colour decreased significantly ($p < 0.05$) in outflow of system with increasing contact time of treatment.
- DO, BOD_5 and COD decreased significantly ($p < 0.05$) in all FTWs.
- The presence of ochre pellets in combination with *P. australis* decreased DO and BOD_5 significantly ($p < 0.05$) and increased COD significantly ($p < 0.05$).
- The DO, BOD_5 and COD values were not affected with increasing contact time in systems using a combination of ochre pellets with *P. australis*. However, BOD_5 and COD increased significantly ($p < 0.05$) with increasing contact time in systems with only *P. australis* and only SGW.
- The concentration of NO_3–N was significantly ($p < 0.05$) higher, and PO_4–P was significantly ($p < 0.05$) lower than those concentrations of inflow, however, there was no significant ($p > 0.05$) change in NH_4–N concentration in FTWs using a combination of ochre pellets with floating *P. australis*.
- The significant ($p < 0.05$) decreases of NH_4–N and PO_4–P concentrations were linked to the effect of the presence of ochre pellets in combination with *P. australis* and with increasing contact time.

- The presence of ochre pellets in combination with *P. australis* significantly ($p < 0.05$) decreased B and Mg (Al, B, Cd, Cr, Cu, Mg, Ni and Zn), while increasing Al, Ca and Fe (Al, Ca, Fe and K) contents in outflow.
- The effects of *P. australis*, when present in combination with ochre pellets, were significant ($p < 0.05$) in decreasing the concentrations of all considered elements in the SGW.
- The presence of floating *P. australis* contributed significantly ($p < 0.05$) to the removal of B, Ca, Cd, Cr, Cu, Fe, Mn, Ni, and Zn.
- Increasing contact time significantly ($p < 0.05$) decreased Al, B and Ni, while it increased Ca, Fe, and Mg significantly ($p < 0.05$) compared with the outflow of the system treating LC–SGW for 2–day with a combination of ochre pellets and floating *P. australis*.
- There were no significant ($p > 0.05$) effects of increasing contact time of treatment of HC–SGW by a combination of ochre pellets with floating *P. australis* on removal of trace elements.
- The removals of Al, Ca, Fe and Na, were higher in systems treating HC–SGW, while removals of Cd, Cu, Mg, Mn, Ni and Zn were higher in systems treating LC–SGW.
- Accumulation of all considered elements was significantly ($p < 0.05$) higher in *P. australis* of all treatment systems (T) than in control systems (C), except for Na which had significantly ($p < 0.05$) lower accumulations.
- The presence of ochre pellets in combination with *P. australis* in wetlands treating SGW significantly ($p < 0.05$) reduced the concentrations of all considered trace elements which accumulated in *P. australis* tissues, except for Ca which increased significantly ($p < 0.05$).

- The concentrations of almost all elements, which accumulated in *P. australis* tissues, were significantly ($p < 0.05$) lower in wetlands treating both types of SGW for 7–day contact time compared to those concentrations which accumulated in wetlands of 2–day contact time treatments. This was inverse to the behaviour exhibited in control wetlands.
- Accumulation rates in tissues of *P. australis* were higher for all considered trace elements in wetlands treating HC–SGW compared to those values in wetlands treating LC–SGW, except for the Ni accumulation rate which was higher in wetlands treating LC–SGW.
- The statistical analysis showed that accumulated concentrations of all considered elements in ochre pellets after the end of the treatment experiment were significantly ($p < 0.05$) higher, except for Ca concentrations which were significantly ($p < 0.05$) lower, than those in ochre pellets before commencing the experiment.
- Presence of *P. australis* in combination with ochre pellets significantly ($p < 0.05$) increased the concentrations of Al, B, Ca, Cr, Cu, Fe and Zn which accumulated in the ochre pellets.
- For HC–SGW, ochre pellets adsorption of trace elements was effective in 2–day treatment time with presence of *P. australis*, in terms of Al, Ca and Cd.
- For LC–SGW, there were significant ($p < 0.05$) increases in accumulated Cd and Cr in ochre pellets with increasing contact time.
- In systems using a combination of ochre pellets and *P. australis*, the concentrations of B, Cd, Cr, Cu and Mg, which accumulated in the ochre pellets of systems treating HC–SGW, were higher than those values in the ochre pellets of systems treating LC–SGW.
- Contents of Al, Ca and Fe which accumulated in ochre pellets were higher in treatment of LC–SGW compared with corresponding systems treating HC–SGW.
- Accumulation of trace elements in rhizomes of *P. australis* were significantly ($p < 0.05$) higher than in stems and leaves.

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Appendix

Published Researches